Validation of version 5.20 ILAS HNO3, CH4, N2O, O3, and NO2 using ground-based measurements at Arrival Heights and Kiruna

S W. Wood, National Institute of Water and Atmospheric Research

G E. Bodeker, National Institute of Water and Atmospheric Research

I S. Boyd, National Institute of Water and Atmospheric Research

Nicholas Jones, University of Wollongong, njones@uow.edu.au

B J. Connor, National Institute of Water and Atmospheric Research

See next page for additional authors

Follow this and additional works at: https://ro.uow.edu.au/smhpapers

Part of the Medicine and Health Sciences Commons, and the Social and Behavioral Sciences Commons

Recommended Citation


Research Online is the open access institutional repository for the University of Wollongong. For further information contact the UOW Library: research-pubs@uow.edu.au
Validation of version 5.20 ILAS HNO3, CH4, N2O, O3, and NO2 using ground-based measurements at Arrival Heights and Kiruna

Abstract
Ground-based measurements of O3 and NO2 columns at Kiruna (68°N) and Arrival Heights (78°S) were used to validate stratospheric profile measurements of the same species by the satellite-based Improved Limb Atmospheric Spectrometer (ILAS) flown between November 1996 and June 1997. In addition, measurements of HNO3, CH4, and N2O made at Arrival Heights are compared with measurements of these species by ILAS. Even though there were no temporal coincidences in the latter comparison, the ground-based data provide a climatological context for the ILAS measurements. ILAS profiles measured within 10° of longitude and 2° of latitude of each site were used. For the species with large tropospheric components (CH4 and N2O), ILAS profiles were integrated upward from 12 km and were compared with ground-based retrievals of partial columns above 12 km. For O3, which is primarily stratospheric but has an appreciable tropospheric component, tropospheric columns (typically to 8–10 km altitude) calculated from ozonesonde climatologies were added to the integrated ILAS profiles to allow comparison with ground-based total column ozone measurements. Tropospheric columns of HNO3 and NO2 were assumed to be sufficiently small so that ILAS profiles integrated above 8 km could be compared directly with ground-based total column measurements.

Keywords
20, 5, version, validation, kiruna, heights, ilas, arrival, hno3, measurements, ground, o3, ch4, n2o, no2, GeoQuest

Disciplines
Medicine and Health Sciences | Social and Behavioral Sciences

Publication Details

Authors

This journal article is available at Research Online: https://ro.uow.edu.au/smhpapers/1978
Validation of version 5.20 ILAS HNO$_3$, CH$_4$, N$_2$O, O$_3$, and NO$_2$ using ground-based measurements at Arrival Heights and Kiruna

National Institute of Water and Atmospheric Research, Lauder, Central Otago, New Zealand

F. J. Murcray
Department of Physics, University of Denver, Denver, Colorado, USA

H. Nakajima and Y. Sasano
National Institute for Environmental Studies, Tsukuba, Japan

Received 2 March 2001; revised 13 July 2001; accepted 16 July 2001; published 27 September 2002.

Ground-based measurements of O$_3$ and NO$_2$ columns at Kiruna (68°N) and Arrival Heights (78°S) were used to validate stratospheric profile measurements of the same species by the satellite-based Improved Limb Atmospheric Spectrometer (ILAS) flown between November 1996 and June 1997. In addition, measurements of HNO$_3$, CH$_4$, and N$_2$O made at Arrival Heights are compared with measurements of these species by ILAS. Even though there were no temporal coincidences in the latter comparison, the ground-based data provide a climatological context for the ILAS measurements. ILAS profiles measured within 10° of longitude and 2° of latitude of each site were used. For the species with large tropospheric components (CH$_4$ and N$_2$O), ILAS profiles were integrated upward from 12 km and were compared with ground-based retrievals of partial columns above 12 km. For O$_3$, which is primarily stratospheric but has an appreciable tropospheric component, tropospheric columns (typically to 8–10 km altitude) calculated from ozonesonde climatologies were added to the integrated ILAS profiles to allow comparison with ground-based total column ozone measurements. Tropospheric columns of HNO$_3$ and NO$_2$ were assumed to be sufficiently small so that ILAS profiles integrated above 8 km could be compared directly with ground-based total column measurements.

INDEX TERMS: 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0394 Atmospheric Composition and Structure: Instruments and techniques


1. Introduction

The Improved Limb Atmospheric Spectrometer (ILAS) was flown on the Advanced Earth Observing Satellite (ADEOS), making profile measurements of a range of trace gases in the high latitudes of both hemispheres from November 1996 to June 1997 [Sasano et al., 1999a; Nakajima et al., 2002]. As part of the intensive ILAS validation campaign [Kanzawa et al., 1997], ground-based measurements of O$_3$ and NO$_2$ were made at Kiruna, in Northern Sweden (67.78°N, 21.10°E). Measurements of these two gases and of HNO$_3$, CH$_4$, and N$_2$O were made at Arrival Heights on Ross Island in Antarctica (77.8°S, 166.6°E). This paper presents a comparison of the ground-based measurements made at these two sites with vertically integrated ILAS version 5.20 profile measurements [Yokota et al., 2002]. In addition, the measurements taken over a longer time period at the two sites are presented to establish a climatological context for the ILAS data and thereby aid intercomparisons of ILAS and UARS climatologies presented in associated papers.

The instruments available at the two sites were different. While fewer species were measured at Kiruna, their coincidence with ILAS measurements was better than at Arrival Heights. At Arrival Heights a wider range of instruments was operated but the number of measurements made during the ILAS measurement period was small. However, the seasonal behavior of the measured gases has been well characterized by subsequent measurements.

1.1. Kiruna

The first National Institute of Water and Atmospheric Research (NIWA) UV/vis spectrometer was installed in...
August 1991 at the Swedish Institute of Space Physics laboratory at Kiruna (Ake Steen principal scientist) to measure NO2 and O3. This system was upgraded in March 1997 to allow additional measurements of OCIO and BrO in the 350 nm region. Daily ozone and temperature profiles from ozonesondes launched at Kiruna during February and March 1997 to validate the ILAS ozone profiles [Sasano et al., 1999b] were used to calculate the air mass factors (AMFs) used to convert the UV/vis slant column densities to vertical column densities [Solomon et al., 1987] for comparison with the integrated ILAS profiles.

1.2. Arrival Heights

[5] A larger range of instruments is operated by NIWA at Arrival Heights, some in collaboration with other institutions. Zenith viewing NO2 spectrometer measurements in the 440 nm region have been made at the site almost continuously since 1982 [Keys and Johnston, 1986]. The wavelength range was extended in 1994 to give better retrievals of ozone. In addition, diode-array spectrometers retrieving OCIO between 400 and 424 nm and BrO between 345 and 360 nm have been operated on a campaign basis from spring 1993 and have been permanently stationed there since 1996 [Kreher et al., 1996]. Since the retrieval technique requires spectra to be recorded at high solar zenith angle (SZA), the most useful measurements are confined to those periods when sunrise and sunset events occur, around the equinoxes. During the winter period (May to July) no measurements are made.

[6] Dobson spectrophotometer 17 has been operated at Arrival Heights since 1988 [Nichol et al., 1996]. Primarily direct Sun measurements in the ultraviolet are used to retrieve total column ozone. On cloudy days, measurements using zenith scattered skylight are made, and during winter (April to August) measurements are only possible using moonlight. The zenith and moonlight measurements are considerably more uncertain than the direct Sun measurements due to the lower light intensity. Although total column ozone measurements are also available from the zenith viewing visible wavelength spectrometers, Dobson spectrophotometer measurements have been used in preference in this study because of the difficulty in accurately calculating AMFs at high SZA when there is large variability in the vertical ozone profile. The Dobson spectrophotometer measurements are made at lower SZA so that this is not a concern.

[7] A Fourier transform spectrometer (FTS) has been operated at Arrival Heights in conjunction with the University of Denver since the late 1980s on a campaign basis and full time since 1991 [Kreher et al., 1996; Keys et al., 1998]. In preparation for the ILAS validation campaign the FTS was upgraded to a Bruker model 120M capable of measuring direct solar radiation at wavelengths from 2 to 13 μm with a maximum optical path difference (MPD) of 257 cm (giving a nominal resolution of 0.0035 cm⁻¹). Prior to that a lower resolution Bomem model DA2 FTS (with MPD of 50 cm) was operated at the site, but this was more limited in the wavelengths it could record. Unfortunately the combination of an interrupted measurement programme following the installation of the new FTS and the premature failure of the ADEOS satellite resulted in very few coincidences between FTS and ILAS measurements. However, the longer measurement series does provide a climatological context for the ILAS measurements.

2. Description of Measurement Techniques

2.1. ILAS

[8] ILAS is a sensor based on solar occultation by infrared spectrometry [Suzuki et al., 1995]. Fourteen profile measurements from 57°N to 73°N and 64°S to 88°S with a vertical resolution of 1.6 km [Hayashida et al., 2000, Appendix] were made each day. HNO3, CH4, N2O, O3, and NO2 mixing ratios were retrieved every 1 km and a 13-km-wide sampling volume has been calculated from the ILAS field of view. The air density-weighted absorption of solar radiation occurs over an effective path length of 200–300 km, depending on the altitude. Therefore, each ILAS measurement data point represents an average value in the sampling volume of 13 km (width) × 1 km (height) × 250 km (length). In comparison, the sampling volume of the ground-based measurements is defined by the light path and so depends on the SZA at the time of the measurement. Measurements made at relatively low SZA (<60°) sample the atmosphere close to the measurement site, but those at high SZA sample the stratosphere at distances from 400 to 800 km from the site. When the ILAS measurements are integrated to produce a vertical column, the larger sampling volume of the profile as a whole, results in some spatial averaging compared with the ground-based measurements. This and the fact that the two measurements can sample in slightly different air masses may account for some of the discrepancies between the two data sets especially in the presence of large tracer gradients.

[9] Validation of many of the ILAS species has already been undertaken, e.g., for O3, Lee et al. [1999] and Sasano et al. [1999]; for NO2, Payan et al. [1999]; for HNO3, Koike et al. [2000]; and for N2O and CH4, H. Kanzawa et al. (Validation and data characteristics of nitrous oxide and methane profiles observed by the Improved Limb Atmospheric Spectrometer (ILAS) and processed with version 5.20 algorithm, submitted to Geophysical Research, 2001).

2.2. Fourier Transform Spectrometer (FTS)

[10] Analysis of the infrared spectra from the FTS was made using the SFIT2 software developed jointly by NIWA, NASA Langley and the University of Denver [Connor et al., 1998]. SFIT2 is a radiative transfer and profile retrieval algorithm for applied to spectra recorded in absorption. Multiple spectral windows may be handled simultaneously. It incorporates a line-by-line calculation of the monochromatic atmospheric spectrum using line parameters from the HITRAN database [Rothman et al., 1998]. Atmospheric ray-tracing and the computation of mass paths are made using the FSCATM algorithm [Gallery et al., 1983]. The instrument response is simulated by Fourier transforming the spectrum, applying apodisation and phase functions, and reverse Fourier transforming. The instrument field-of-view is always included in the response function; in addition there is an option to incorporate arbitrary apodisation and phase functions derived from a measured instrument line shape. The retrieval algorithm is optimal estimation [Rodgers, 1976, 1990] with Newtonian iteration to account for...
nonlinearity of the spectral calculation. The retrieved state vector consists of one or more vertical gas profiles, optional scale factors which are applied to profiles of interfering gases, optional instrument parameters including slope, wavelength shift, apodisation and phase parameters, and optional parameters for a solar CO model. A priori values and estimated uncertainties for fitted parameters and for each gas profile are input into the retrieval along with an estimate of the RMS noise in the measurement.

While SFIT2 is capable of retrieving vertical profile information from the observed shape of absorption lines in the spectra, the vertical resolution is limited. For this reason results are presented as either total columns or as partial columns, the latter being contributions to the total columns over specified altitude ranges. An assessment of the information available in the measurement can be made by calculating averaging kernels which demonstrate the response of the retrieved profile, or specified functions of it such as the partial and total columns, to assumed changes in the true atmosphere. This shows that, even where total columns are reported, the use of the profile retrieval technique gives more reliable and accurate results than earlier techniques that simply scaled an assumed vertical profile of the measured gas.

For HNO₃ which is primarily stratospheric, total column amounts were retrieved from the FTS measurements over the wave number range 867.5–870 cm⁻¹. Wave number ranges used for CH₄ and N₂O were 2650.75–2651.3 cm⁻¹ and 2481.2–2483.5 cm⁻¹, respectively. Since ILAS cannot measure below the tropopause, and these two gases have a significant tropospheric component, SFIT2 was used to retrieve a stratospheric partial column (12–100 km) for comparison with integrated ILAS profiles (12–50 km). The choice of 12 km as a cut off altitude was made so that it was above the bottom altitude of all ILAS measurements. The concentration of these gases above 50 km is sufficiently low (less than 0.1% of the total column in both cases) that the difference in the upper limit of integration will not affect these intercomparisons. The stratospheric partial columns represent approximately 12–15% and 8–14% of the total columns of CH₄ and N₂O, respectively. To demonstrate the ability to separate these partial columns from the ground-based measurements, averaging kernel functions, which show the sensitivity of the retrieved partial columns to changes in the atmosphere, as a function of altitude, have been calculated (Figure 1). An ideal averaging kernel would have a value of 1.0 in the region of interest and zero elsewhere. The kernels shown in Figure 1 show that the tropospheric and stratospheric partial columns for CH₄ and N₂O can be separated but that the separation is not perfect and occurs over a scale of a few kilometers. Note also that the sum of the two kernels shown should be equal to 1.0, and that the places where it is not, e.g., for CH₄ near the ground and N₂O near 50 km, it indicates that there is insufficient information in the measurement to completely determine the gas mixing ratio.

An error analysis for the FTS retrieval process was carried out by estimating the errors in the retrieved partial columns from estimates of the uncertainties in a priori information and in the measurement itself following the method of Rodgers [1990]. This calculation showed that the stratospheric partial columns calculated for CH₄ had a higher uncertainty (≈6 × 10¹⁷ molec. cm⁻² or ≈12%) than those for N₂O (≈2 × 10¹⁶ molec. cm⁻² or ≈3%). This is a reflection of the spectroscopy of the absorption lines used in the analysis. Work is under way to improve the vertical accuracy of these retrievals by using multiple wavelength windows. The total columns of HNO₃ had typical uncer-
tainties ranging from $\sim 5 \times 10^{14}$ molec. cm$^{-2}$ for the high resolution measurements to $\sim 8 \times 10^{14}$ molec. cm$^{-2}$ for the lower resolution measurements.

2.3. UV/vis Spectrometer

[14] NO$_2$ and O$_3$ measurements were made using zenith viewing spectrometers measuring scattered sunlight in the 440 nm region of the spectrum at twilight ($75^\circ \leq$ SZA $\leq 95^\circ$). The differential optical absorption spectroscopy (DOAS) technique also requires the measurement of a low SZA reference spectrum to enable the removal of the strong Fraunhofer absorptions present in the extra-terrestrial spectrum by ratioing the twilight measurements with the low SZA measurements. During the twilight period the measured scattered light passes through a long path in the stratosphere enhancing the weak absorptions by stratospheric NO$_2$ and O$_3$ by factors of 5 to 30. Analysis of NO$_2$ was made over the wavelength interval 437–469 nm. For ozone retrievals this was extended to 437–489 nm. Laboratory measured NO$_2$ and O$_3$ absorption cross-sections are least squares fitted to the absorption spectra to calculate slant columns. A discussion of the measurement and retrieval process is given by Kreher et al. [1995]. Vertical column amounts are derived using AMFs calculated using radiative transfer models [Preston et al., 1998]. For NO$_2$, a constant AMF, derived from measurements at mid-latitudes, was used at both sites. The error introduced by this assumption can be as much as 8%, because in reality the AMF will vary by this much with changing temperature and the changing vertical profiles of both NO$_2$ and O$_3$. At Kiruna, ozonesonde profiles from the period of intensive measurements at Kiruna in early 1997 were used to calculate daily AMFs for O$_3$. Representative values of these AMFs were used for the measurements at other times. This greatly reduces the uncertainty in the conversion of the measurements to vertical column densities.

[15] The analysis of measured spectra to produce slant column amounts requires techniques that include corrections for instrumental and atmospheric artifacts. In the data presented here, this included:

1. nonlinear least squares fitting to correct for spectral shift and stretch; spectra are accurately aligned to the Kitt Peak solar atlas [Kurucz et al., 1984] after convolving them with the instrument function,
2. convolution of all fitted cross-sections with the instrument function before fitting,
3. use of low temperature (227K) NO$_2$ absorption cross-sections [Harder et al., 1997],
4. use of low temperature (241K) O$_3$ absorption cross-sections (University of Bremen, GOME cross-sections)
5. use of cross-sections for all known absorbers in the 435–490 nm region: NO$_2$, O$_3$, H$_2$O, and O$_4$,
6. correction and fitting for the effect of rotational Raman scattered light (Ring effect),
7. use of filters to minimize the effect of Rayleigh and Mie scattering wavelength dependent functions present in scattered light spectra,
8. correction of cross-section shape to allow for the effect of the Fraunhofer structure in the light source.

[16] Examples of the low errors that can be achieved with this analysis are given by Harder et al. [1997]. In particular, the use of absorption cross-section data for temperatures close to stratospheric temperatures is a significant improvement on using room temperature cross-sections. However, there can still be systematic errors of up to 5% in retrieved columns because real atmospheric temperatures will vary. A further discussion of the errors in both the retrievals and the calculation of air mass factors in the context of polar measurements is given by Sanders et al. [1999].

2.4. Dobson Spectrophotometer

[17] Total column ozone amounts are derived from Dobson spectrophotometer measurements of the ratio of the solar irradiance at two different ultraviolet wavelengths. Monthly lamp calibrations are made to detect instrumental drifts and periodic intercomparisons are made against a regional standard Dobson instrument in Australia. These intercomparisons were carried out in January 1996 and January 1999. The error in the most accurate direct-sun Dobson spectrophotometer measurement using a well-calibrated and operated instrument is about 1% (1$\sigma$) [Basher, 1995] and the potential long-term stability is about ±0.5% (1$\sigma$) for annual means [World Meteorological Organisation (WMO), 1980]. Zenith cloudy measurements made at high SZA were found to be biased when compared to direct-sun measurements made on the same days and so an empirical correction, calculated from the days when both measurement types were available, was applied to the cloudy measurements on days when that was the only measurement type made.

3. Data Selection and Preprocessing

[18] ILAS version 5.20 profile measurements within 2$^\circ$ of latitude and 10$^\circ$ of longitude of Arrival Heights and Kiruna were used in this validation study. Since the latitude of the ILAS measurements changes rapidly in the southern hemisphere [Sasano et al., 1999a], the 2$^\circ$ latitude constraint limited the number of ILAS profiles available for comparison with the ground-based measurements at Arrival Heights.

[19] The ILAS vertical profiles were integrated using coincident UKMO temperature and pressure profiles to derive column amounts which could then be compared with the ground-based measurements. Uncertainties in the column amounts were calculated using the root sum square of the uncertainties at each altitude level. This is a conservative calculation of the uncertainties since the measurements at each level are not completely independent. Since ILAS profiles do not extend into the troposphere, there is an offset between the ILAS and ground-based measurements for any species with an appreciable tropospheric component, e.g., O$_3$. Accordingly, a tropospheric integrated ozone profile climatology was calculated from the February and March ozonesonde flights at Kiruna (integrated ozone profiles up to 8.5–14.5 km in 1-km steps were 25.9 ± 5.2 (1$\sigma$), 30.2 ± 5.9, 37.3 ± 8.0, 47.5 ± 11.1, 60.5 ± 14.6, 75.2 ± 18.0, and 92.1 ± 22.1 DU) and were added to the Kiruna ILAS ozone profiles below their minimum altitude. Likewise, for the southern hemisphere, 506 ozoneonde flights made at McMurdo (2 km from Arrival Heights) from 1986 to 1998 were used to calculate a tropospheric integrated ozone profile climatology (integrated ozone profiles up to 8.5–14.5 km in 1-km steps were 18.7 ± 5.4 (1$\sigma$), 21.2 ± 5.7, 25.6 ± 6.3, 32.1 ± 7.3, 39.2 ± 8.5, 45.9 ± 10.3, 52.6 ± 13.6
DU) which was added to the ILAS ozone profiles below their minimum altitude.

[20] For NO₂ and HNO₃ no similar correction was made to the integrated ILAS profiles as tropospheric components were assumed to be negligible. Remote, free tropospheric HNO₃ mixing ratios are typically around 100 pptv [LeBel et al., 1990] which contribute \( \sim 7.7 \times 10^{14} \) molec. cm⁻² to the total column (\( \sim 5\% \)). Tropospheric NO₂ concentrations are very low in the remote, clean, free troposphere, with summertime tropospheric columns almost always less than \( 0.1 \times 10^{16} \) molec. cm⁻² [Velders et al., 2001] which is \( \sim 15\% \) of the column, but are significantly higher in polluted air, e.g., following biomass burning. At Arrival Heights the troposphere will always be clean but at Kiruna episodic tropospheric pollution events may affect the ground-based measurements, as discussed below. For CH₄ and N₂O, a ground-based column measurement would be dominated by the tropospheric component which is not measured by ILAS. Therefore, for these gases the SFIT2 retrieval was used to separate the tropospheric and stratospheric parts of the total column at approximately 12 km altitude. ILAS profiles were then integrated from 12 to 50 km, even if the profile extended below 12 km, to match the SFIT2 retrieval.

4. Intercomparison Results

4.1. Kiruna NO₂ and O₃

[21] The ground-based measurements of NO₂ at Kiruna through 1996 and 1997 are shown in Figure 2. Sunrise and sunset measurements are plotted separately to show the diurnal variation which maximizes at the equinoxes. The time series for three different SZAs are also shown to indicate the precision and repeatability of the measurements. The fact that these three curves cannot always be distinguished shows that the AMF calculations for the different SZAs are consistent and that random errors are relatively small. However, systematic errors of up to 14% can result from variation in the temperature of the absorbing gas and in the AMF. The occasional upward spikes in the NO₂ vertical columns arise from episodic tropospheric pollution events at Kiruna. ILAS and ground-based NO₂ column measurements are shown during their period of overlap in Figure 3. Only the evening ground-based measurements are shown since they are closest in time to the ILAS measurements made in the Northern Hemisphere. The agreement between the ILAS and ground-based measurements is good and most often within the uncertainty in the ILAS data. The average bias of the ILAS measurements, relative to a daily average of the sunset ground-based measurements is +5.4% but the standard deviation of this result (1σ) is 20%.

[22] Total column ozone measurements from ILAS and the UV/vis spectrometer for all of 1996 and 1997 are shown in Figure 4. Only the evening ground-based measurements are shown. As noted in section 2, the use of daily AMFs means that the errors from incorrect AMF should be much smaller than for NO₂, where fixed AMF values were used. As in earlier figures, the measurements from three different SZAs are plotted to show the consistency of the retrievals. In most cases these curves are indistinguishable; places where they differ appreciably indicate either real temporal changes between the measurements or inconsistencies in the measurements or AMF calculations. Total column ozone levels show large fluctuations as Kiruna moves into and out of the polar vortex. The integrated ILAS profiles generally
agree well with the ground-based measurements and track these fluctuations well. The period of coincident measurements during 1997 is shown in more detail in Figure 5. Note that the January measurements at the left of this figure represent a time when the Sun is only just returning to the site and the differences between measurements made at different zenith angles are much larger because of a larger separation in time and viewing direction, and the possibility

Figure 3. Detail of the coincident 1997 NO₂ measurements at Kiruna. Sunset measurements from the ground-based measurements at 88°, 90°, and 92° SZA are shown using solid lines. Vertical columns of NO₂ calculated from the ILAS v5.20 profiles are shown using diamonds together with their 1σ uncertainties.

Figure 4. Ground-based measurements of O₃ at Kiruna for 1996 and 1997. Sunset measurements at 88°, 90°, and 92° SZA, each converted to a vertical column density using the appropriate air mass factors, are plotted in black and shades of grey. Vertical ozone columns from ILAS v5.20 profile measurements are shown using diamonds together with their 1σ uncertainties.
of real air mass or ozone changes between measurements on the same day. The average bias of the ILAS measurements relative to a daily average of the sunset ground-based measurements is \(-1.7\%\) with a standard deviation of 10\%, despite variations in the total column ozone of \(\pm 15\%\). The ILAS measurements track most of the ground-based measurements well though they tend to show less variability, i.e., the extreme values are not reproduced. This may be a result of the larger sampling volume of the ILAS measurement which, in the presence of large spatial gradients in ozone, would tend to smooth the data. There is one period of approximately a week in mid-March where the integrated ILAS profiles appear to differ significantly from the ground-based measurements. A close examination of this period (not presented here) showed that the morning ground-based measurements, which look eastward, were lower than the evening, westward looking, measurements (which are plotted in Figure 5) by as much as 15\% on some of the days, and were actually much closer to the values calculated from integrated ILAS profiles. Strong east-west gradients in total column ozone in the vicinity of Kiruna, were observed in TOMS data over this one week period and account for the AM–PM asymmetry in the ground-based measurements. A calculation of isentropic potential vorticity (IPV) from UKMO temperature and wind data confirmed that there were strong horizontal gradients in IPV, associated with the edge of the polar vortex, in the region of Kiruna, during this period.

4.2. Arrival Heights NO\textsubscript{2} and O\textsubscript{3}

[23] Ground-based measurements of NO\textsubscript{2} columns at Arrival Heights during 1996 and 1997 are shown in Figure 6 together with the integrated ILAS profiles satisfying the selection criteria. The nature of the ILAS latitudinal coverage in the southern hemisphere [Sasano et al., 1999a] results in two brief periods when ILAS measurements were made within 2\° of Arrival Heights: from 20 to 27 February 1997 (P1) and 16 to 24 April 1997 (P2). During P1 the ILAS measurements are approximately 40\% higher than the ground-based measurements while during P2 the ILAS measurements are considerably closer to the ground-based measurements.

[24] Total column ozone measurements made with Dobson spectrophotometer 17 at Arrival Heights are shown together with integrated ILAS ozone profiles in Figure 7. The nature of the latitudinal coverage of the ILAS data and the fact that the Dobson spectrophotometer does not make measurements at high SZA, results in a short period of overlap between the two data series from 20 February to 1 March 1997. During the overlap period, the integrated ILAS profiles are 5 to 8\% higher than the ground-based measurements.

4.3. Arrival Heights HNO\textsubscript{3}, CH\textsubscript{4}, and N\textsubscript{2}O

[25] Ground-based measurements of total column HNO\textsubscript{3} for the 3 years from mid-1996 to mid-1999 are shown in Figure 8 together with integrated ILAS nitric acid profiles (only four profiles matched the selection criteria). The bulk of the ground-based measurements were made with the Bruker FTS using sunlight. Measurements in 1996 were made with the lower resolution Bomem FTS. In 1998, some measurements using moonlight were made and these are also shown. While there is no precise temporal coincidence between the ILAS and ground-based measurements, the ILAS data do follow the seasonal increase observed in the
ground-based measurements and appear to be a little lower than what could be expected from the ground-based measurements. The likely reason for this is that the comparison ignored the ∼5% tropospheric component of the HNO₃ column which is not included in the integrated ILAS profiles.

[26] The total CH₄ column from the ground-based measurements is approximately 3.3 × 10¹⁹ molec. cm⁻² and that
of N$_2$O is $5.9 \times 10^{18}$ molec. cm$^{-2}$. The ability to separate a stratospheric component of these columns allows a more meaningful comparison with integrated ILAS profiles. For these molecules the ground-based measurements presented are partial columns from 12 km upward as discussed in section 2. Comparisons of ground-based partial columns and integrated ILAS profiles for CH$_4$ and N$_2$O are shown in Figure 9 and Figure 10, respectively. While no coincident ground-based and ILAS CH$_4$ measurements are available, the integrated ILAS profiles agree with what could be expected from the ground-based measurements within their combined uncertainties. However, the ILAS measurements do not capture the prewinter decrease seen in the ground-based measurements during 1998 and early 1999. The prewinter decrease results from diabatic descent inside the polar region which brings CH$_4$ poor air from higher to lower altitudes. Likewise there are no coincident ground-based and ILAS N$_2$O measurements. However, the ILAS N$_2$O measurements are in approximate agreement with what could be expected from the ground-based measurements and track the prewinter decrease in N$_2$O observed in the 1998 ground-based measurements.

5. Discussion and Conclusions

[27] This paper has presented a comparison of ILAS measurements with a variety of ground-based measurements with quite different characteristics. Some of these measurements have no sensitivity at all to the vertical distribution of the measured gas and others do have limited vertical resolution. To adopt a uniform approach in comparisons between the ILAS measurements and ground-based measurements containing different vertical distribution information, total columns have been compared where possible. If there is a known mismatch in the vertical range of sensitivity that would introduce an offset in this comparison, techniques to eliminate it by either adding an estimate of the “missing” contribution to the total column, or by comparing partial columns instead, have been used. A more detailed comparison would look at the vertical sensitivities of the two measurement systems and convolve the retrievals from one instrument with the sensitivity function of the other. This approach is described by Rodgers and Connor [1999]. Since we are not attempting to compare vertical profiles from different measurements in this work, we have assumed that the simpler method of comparing total or partial columns is sufficient to provide a meaningful comparison.

[28] At Kiruna, the integrated ILAS NO$_2$ profiles agree very well with the ground-based measurements. The agreement between integrated ILAS O$_3$ and ground-based total column ozone measurements is a little poorer, but there is evidence that this is quite likely due to the high spatial variability of ozone during the Arctic spring and the fact that the sampling volumes of the two measurements are different.

[29] Although a larger array of instrumentation is available at Arrival Heights compared to Kiruna, the rapid latitudinal change in ILAS coverage and the 2° latitude selection constraint used here, results in fewer coincidences between ground and satellite-based measurements at Arrival Heights than at Kiruna. This limits the conclusions that can be drawn from the comparisons at Arrival Heights. While integrated ILAS ozone profiles agree reasonably well with the ground-based total column ozone measurements, it is clear that NO$_2$ values derived from ILAS have a larger

---

**Figure 8.** Total column HNO$_3$ measurements at Arrival Heights from ILAS (diamonds with 1σ uncertainties) and ground-based FTS (various symbols detailed in legend). In 1998 some successful lunar measurements were made with the new Bruker FTS.
annual cycle amplitude than is seen in the ground-based measurements. For other species the ILAS measurements were broadly consistent with the seasonal behavior observed in ground-based measurements.

[30] Although the spatial selection constraints used here are quite typical for comparisons of ground and satellite-based measurements, they are not necessarily the most physically relevant criteria. A future extension of this work

Figure 9. Ground-based measurements of partial columns of CH$_4$ from 12 to 100 km (filled circles) together integrated ILAS CH$_4$ profiles from 12 to 50 km (diamonds with 1σ uncertainties). The stratospheric column derived from the ground-based measurements is between 12% and 15% of the total column.

Figure 10. Measurements of N$_2$O at Arrival Heights from 1997 to 1999. As for Figure 9, these values are partial columns only, which is between 8% and 14% of the total column. ILAS v5.20 measurements are shown as in previous figures.
will use backward and forward trajectories from the ILAS measurement locations to extend the spatial coverage of the ILAS data for long-lived species such as N₂O and CH₄. This is expected to considerably increase the coincidences between satellite and ground-based measurements for these species. Future comparisons between ground-based and satellite-based measurements will also be enhanced as we refine the measurement and retrieval techniques to increase vertical distribution information from the ground-based measurements.

Acknowledgments. Measurements at Arrival Heights were made with logistic and technical support from Antarctica New Zealand. The University of Denver assisted with support of the Arrival Heights FTS measurements. The measurements made at Arrival Heights and Kiruna were funded in part by the ILAS funded validation experiment “Coordinated ground-based observations at high latitudes” (Andrew Matthews PI). Greg Boderke was supported by ISPS funding for a 1-year postdoctoral position in Japan to work on the ILAS program. The analysis of the ILAS and ground-based measurements used in this study was undertaken within the New Zealand FRST funded programs CO1X033 and CO1X035. We thank Terry Deshler for providing the McMurdo ozonesonde data, through the NDSC, used in this study.

References


