Assessment of Improved Limb Atmospheric Spectrometer (ILAS) Version 6 data quality: measurements of stratospheric O₃, HNO₃, NO₂, N₂O, CH₄, H₂O, and aerosol extinction coefficient

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Abstract. The Improved Limb Atmospheric Spectrometer (ILAS) aboard the Advanced Earth Observing Satellite (ADEOS) observed vertical profiles of ozone (O₃), nitric acid (HNO₃), nitrogen dioxide (NO₂), nitrous oxide (N₂O), methane (CH₄), water vapor (H₂O), and other several gaseous species such as chlorine nitrate (CIONO₂) and dichlorodifluoromethane (CF₂Cl₂, CFC-12), as well as aerosol extinction coefficient at 780 nm in the high-latitude stratospheres from November 1996 through June 1997. These data processed with the Version 6 retrieval algorithm are compared with those obtained by balloon-borne and satellite-borne sensors. Through these comparisons, data quality of the Version 6 is evaluated. The quality of the Version 6 data set is generally comparable to that evaluated for the former Version 5.20, which was well validated [*Sasano*, 2002; and references therein] and available to the general public. CIONO₂ and CFC-12 data are newly added to the Version 6 data set. The Version 6 data set includes more scenes (roughly 300) and lower detectable altitudes (down to 7 km) compared to the Version 5.20 data set.

1. Introduction

It is essential to monitor and understand current trends in the vertical distribution of stratospheric chemical species such as ozone (O₃), aerosol, water vapor (H₂O), reactive nitrogen, and others in order to predict their future trends. The abundances of such species in the upper troposphere and stratosphere are important components of the stratospheric climate system that directly influence ozone [e.g., *WMO*, 2003]. Satellite-borne sensors such as the Stratospheric Aerosol and Gas Experiment (SAGE) I and II, the Halogen Occultation Experiment (HALOE), the Microwave Limb Sounder (MLS), the Polar Ozone and Aerosol Measurement (POAM) II and III, as well as in situ or remote-sensing sensors from the air or ground contribute to current understanding of long term changes in these species [e.g., *SPARC*, 1998; 2000]. Moreover, several atmospheric measurement missions from satellites has successfully started under international cooperation since 2001.

In order to monitor the changes in distributions of stratospheric ozone and its related species in the high latitudes, the Improved Limb Atmospheric Spectrometer (ILAS), which makes use of the solar occultation technique, aboard the Advanced Earth Observing Satellite (ADEOS) was launched from Tanegashima island, Japan (30°N, 131°E) in August 1996. Until June 30, 1997, when the ADEOS stopped working owing to a solar paddle array failure, about 5800 vertical profiles were successfully retrieved so far [*Sasano et al.*, 1999]. ILAS filled a gap between the Polar Ozone and Aerosol Measurement (POAM) II and III measurement periods (October 1993 to November 1996 for POAM II and April 1998 to the present for POAM III) [*Bevilacqua*, 1997; *Lucke et al.*, 1999]. POAM also makes use of the solar occultation technique.

ILAS instrument is described by *Nakajima et al.* [2002a], and ILAS Version 5.20 algorithm is described by *Yokota et al.* [2002] and *Nakajima et al.* [2002b]. ILAS Version 5.20 data quality is generally very good based on validation analyses. The data quality depends on the parameters of the Version 5.20 product, and the details are not described here, but can be found in the cited references below. On the basis of comprehensive comparison of ILAS Version 5.20 data with various sources of validation data, ILAS O₃ validation analysis was carried out by *Sugita et al.* [2002], HNO₃ and NO₂ by *Irie et al.* [2002], H₂O by *Kanzawa et al.* [2002, 2003a] and *Pan et al.* [2002], N₂O and CH₄ by *Kanzawa et al.* [2003b], aerosol extinction coefficient (AEC) at 780 nm by *Saitoh et al.* [2002]. Validation analyses on multiple species were carried out by *Jucks et al.* [2002] and *Toon et al.* [2002] on the basis of comparison of LAS Version 5.20 data with balloon experiment data at Fairbanks. These papers were published in ILAS Special Section of Journal of Geophysical Research in December 2002. The Version 5.20 data set is available to the general public through http://www-ilas.nies.go.jp/.

This report focuses on an assessment of the quality of the ILAS O₃, HNO₃, NO₂, N₂O, CH₄, H₂O, and AEC at 780 nm data processed with the retrieval algorithm, Version 6, through comparisons with other established measurements. The Version 6 data use several updated information on molecular spectroscopic data and others, which was not considered in the Version 5.20. In addition, about 300 more scenes have been added to the Version 6 data set. The quality of the other Version 6 species such as chlorine nitrate (ClONO₂) and dichlorodifluoromethane (CF₂Cl₂, CFC-12) is separately evaluated by *Nakajima et al.* [2005] and Khosrawi et al. [2004], respectively. During the ILAS measurement period, November 1996 through June 1997, satellite-borne solar occultation sensors were operated: SAGE II [Mauldin et al., 1985] which has been operating since October 1984 and the Halogen Occultation Experiment (HALOE) [Russell et al., 1993] which has been operating since October 1991. We used these two data sets for validating the ILAS data. Data obtained by remote-sensing or in situ sensors from balloons during the ADEOS/ILAS validation campaign conducted in Kiruna, Sweden (68°N, 21°E) and Fairbanks, Alaska (65°N, 148°W) [Kanzawa et al., 1997] are also used. These balloon data are also available to the general public through http://www-ilas.nies.go.jp/. In this paper, the HALOE data

are used for validating O_3 , CH_4 , and H_2O , the SAGE II data for AEC, and the balloon data for the rest of the gaseous species (HNO₃, NO₂, and N₂O).

2. Instrumentation and data sets

2.1. ILAS

ILAS is a solar occultation sensor which consists of two grating spectrometers (covering 6.21-11.77 μ m with a 44-spectral element pyroelectric array detector and 0.753-0.784 μ m with a 1024-spectral element metal-oxide-semiconductor (MOS) photodiode array detector, respectively) and a sun edge sensor [*Nakajima et al.*, 2002a; *Sasano et al.*, 1999]. Vertical profiles of O₃, HNO₃, NO₂, N₂O, CH₄, and H₂O are obtained from the infrared spectrometer, while AEC at 780 nm is obtained from the visible/near-IR spectrometer. The sun-edge sensor is used for determining a tangent height (TH) for each limb measurement.

The instantaneous field of view (IFOV) at a TH has a 1.6 km (1.6 km) width in the vertical and a 13 km (2.0 km) width in the horizontal direction for the IR (visible/near-IR) spectrometer. The partial slant path along the line of sight within a 1 km thick layer just above the TH of 20 km is less than 230 km. With a sampling rate of 12 Hz, a full spectrum over the 44 IR spectral elements is acquired within 0.88 msec, i.e., one major frame (a limb measurement) is corresponding to about 110 m at the TH of 15 km and 270 m at the TH of 55 km, depending on atmospheric refraction. Time-series smoothing, which corresponds to about 10 major frames, is applied in the transmittance data, so the actual vertical resolution is 1.9 km at the TH of 15 km and 3.5 km at the TH of 55 km [*Yokota et al.*, 2002].

The ADEOS satellite was put into a sun-synchronous polar orbit. The inclination angle of ADEOS is 98.6°, and the equator crossing time is around 10:40 local mean solar time (descending). Therefore, the ILAS occultation event occurred at sunrise and sunset seen from the ADEOS satellite on each of about 14 orbits per 24 hours. The measurement region of the ILAS is over high latitude (57-73°N and 64-88°S).

Vertical profiling of atmospheric constituents is performed by using an 'onion-peeling' method. The retrieved altitude, for example for ozone, ranges from 7 km (at the lowest) to 70 km. A detailed description for the retrieval is given by *Yokota et al.* [2002]. A summary of error analysis for the Version 6 ILAS data is shown in Tables 1 and 2. The error values of the Version 6 data are sorted into 'internal' and 'external'. 'Internal' errors refer to errors calculated from the final residuals after convergence of the non-linear least squares fitting for observed and simulated transmittances. 'External' errors refer to errors associated with the calculation of simulated transmittance through uncertainties in the non-gaseous component correction and temperature profiles, which are used as inputs for the retrieval.

In the Version 6, the non-gaseous component correction is still required in order to derive vertical profiles of the gaseous concentration in the altitude range where extinction due to aerosol particles (sulfate aerosols and/or PSCs) can not be neglected [*Yokota et al.*, 2002]. To determine the non-gaseous component in the simulated transmittance, we first evaluate the optical depth due to the gaseous component at the 4 spectral elements where the absorption due to gaseous species is relatively small (so called 'window spectral element'). To accomplish this, we use profiles for O₃, HNO₃, NO₂, N₂O, H₂O, CH₄, CO₂, and other minor gaseous concentrations (such as CFC-11) from an ILAS reference atmosphere model [*Yokota et al.*, 2002]. Then, the non-aerosol (i.e., gaseous only) optical depth at all the other 40 spectral elements is calculated by linear interpolation between these 4 window spectral elements. Use of data from this reference atmosphere model would make some errors for the calculation of simulated transmittance, and considered as the external error. The interpolation would also produce systematic errors for the retrieved profiles of the gaseous concentration, but we do not include it to the external error. We evaluate the corresponding uncertainty below in this subsection.

Effects of uncertainties in temperature, which are used in the calculation of simulated

transmittance, are also included (the uncertainties of ± 2 K at 10 km altitude and ± 5 K at 70 km altitude are assumed for the U. K. Meteorological Office (UKMO) stratospheric analyses [*Swinbank and O'Neil*, 1994], which is used for the input of the ILAS retrieval algorithm) as the external error. A detailed description of error analysis is given by *Yokota et al.* [2002]. The estimated root-sum-square (RSS) total error (internal + external errors) in mixing ratio has been converted in percent error by using each of the retrieved value. Then the median value of individual relative errors for each hemisphere and each species are listed in Table 1a and 1b, respectively, and shown in Figure 1a and 1b, respectively. We used this total error provided in each Version 6 ILAS data file for comparisons/discussion below. However, after the internal release of the Version 6 data set for the ILAS science team members, some modifications to the calculation of the internal error was made (see Appendix 1), which has led to Version 6.1 data set generation.

In addition, relative standard deviation (RSD) was calculated as one sigma standard deviation (around the mean) divided by the mean value over the most quiescent period (roughly 7-8 days duration) for each altitude, instead of giving quiescent periods in advance as was done in *Yokota et al.* [2002] and *Khosrawi et al.* [2004]. Although RSD values for the Version 6 CFC-12 data are shown in *Khosrawi et al.* [2004], it was re-calculated here and shown in Table 1 and Figure 1. In order to search for the most quiescent period, RSD was calculated for consecutive one hundred occultation events at every fifty occultation events from November 1996 through June 1997 for both of the hemispheres. Then, we discarded the RSDs for which the available data numbers are less than 30 out of 100 at respective altitudes. The smallest RSDs, which are considered to be the most quiescent periods, were generally found from January and February 1997 in the Southern Hemisphere (SH) or June 1997 in the Northern Hemisphere (NH). This quantity represents a kind of repeatability of the measurement, which may include a real geophysical variability, giving an upper limit of the repeatability. The RSD values thus determined were generally

smaller than the RSS total error defined above for all of the species (see Table 1 and Figure 1). These results are in consistent with those of the Version 5.20 data set [*Yokota et al.*, 2002] and the Version 6 CFC-12 data [*Khosrawi et al.*, 2004].

Two points should be noted regarding uncertainties in the retrieved profiles. First, it is crucial whether TH is determined accurately or not, because the TH ambiguity propagates directly to the uncertainty in the retrieved volume mixing ratio profiles. For the Version 5.20 algorithm, the TH was determined with a method discussed by *Nakajima et al.* [2002b]. The estimated uncertainty in the altitude registration for the tangent point is 300 ± 360 m. Using the Version 6 retrieval algorithm, the systematic error of 300 m has been reduced significantly owing to the use of the High-resolution Transmission (HITRAN) 2000 database (see Section 3.1.1), while the random error of 360 m still remains.

The second point is the systematic errors associated with the non-gaseous component correction by the simple linear interpolation between the window spectral elements. То evaluate them, we simulated transmittances for cases with several types of IR absorption spectra for sulfate aerosols (50 and 75 wt % H₂SO₄/H₂O binary solutions) and PSCs (nitric acid trihydrate (NAT), supercooled ternary solutions (STS) for four different compositions, and ice) as non-gaseous component and with the above-mentioned reference profiles (a priori profiles) as gaseous components [Yokota et al., 2002]. Using these simulated transmittances, retrievals of the vertical profile of the gaseous concentration were made with applying the linear interpolation method for the non-gaseous contribution in the Version 6 retrieval algorithm discussed here. The difference between each set of the *a priori* and the retrieved gas profiles expressed in number density were well correlated with AEC at 780 nm. Therefore, we can evaluate the systematic errors in the gas number density as a function of the AEC at 780 nm. Assuming typical air number densities at altitudes of 15, 20, and 25 km, the systematic errors in terms of volume mixing ratio are listed in Table 2 for two compositions of sulfate aerosol and for each of the gaseous species. Systematic errors for

the PSC cases are also listed in Table 2. Detailed description of the bias error analysis is given by *Yokota et al.* [2002]. Figures 2a to 2f show these bias errors, in the same manner as Figure 7 of *Yokota et al.* [2002].

2.2. SAGE II and HALOE

In this subsection, we used the SAGE II Version 6.1 data (for validating AEC) and the HALOE Version 19 data (for validating O₃, CH₄, and H₂O). All the data are available for scientific use through their World Wide Web servers (see Acknowledgement). For Version 6.0 SAGE II AEC data, the accuracy is estimated to be within 5-20% [*Hervig and Deshler*, 2002]. Since there are no validation results on the Version 6.1 SAGE II AEC data, we have compared the SAGE II Version 6.0 and Version 6.1 AEC data for the ILAS measurement period. The difference between the two SAGE II data versions is as small as 5% below 25 km. In this analysis, however, we only used random error values described in each SAGE II data file.

Comparative studies using the Version 19 HALOE ozone data with other satellite-borne ozone data are given elsewhere [e.g., *Danilin et al.*, 2002; *Manney et al.*, 2001; *Morris et al.*, 2002]. Those results suggest that they generally agree to each other within 0.2 to 0.5 ppmv (or 4-12%) in most of the stratosphere. For error values of the Version 19 HALOE ozone data, we used the RSS of errors associated with the aerosol effects, which are described in each HALOE data file, and errors given in Table 1 of *Brühl et al.* [1996] except for errors associated with the aerosol effects (*James M. Russell III*, private communication, 2000).

A comparative study of H_2O profiles measured by *in situ* or remote-sensing instruments from aircraft or balloon and several satellite instruments, including the Version 19 HALOE data, was made by *Michelsen et al.* [2002]. The result suggests that H_2O data generally agree to each other within 15% in the lower and middle stratosphere. Although there is no comprehensive comparisons of the Version 19 HALOE CH₄ data with other instruments, *Michelsen et al.* [2002] also demonstrated good agreement with the Version 3 Atmospheric Trace Molecule Spectroscopy Experiment (ATMOS) CH_4 data. For error values of the Version 19 HALOE H₂O and CH_4 data, we also used the RSS of errors associated with the aerosol effects, which are described in each HALOE data file, and errors given by *Harries et al.* [1996] and *Park et al.* [1996], respectively, except for errors associated with the aerosol effects.

2.3. Balloon-borne instruments

Data from balloon-borne instruments are also used for the validation of ILAS Version 6 HNO₃, NO₂, and N₂O. These data have been archived in the ILAS-Correlative Measurement Database (CMDB), which is available to the general public with some restrictions (http://www-ilas.nies.go.jp). Observation date and time, launch site, location (latitude and longitude), vertical resolution, target species used for this study, and principal investigators of each instrument are summarized in Table 3. Here we briefly describe the instrumentation of each sensor used for the ILAS/ADEOS validation campaigns conducted in Esrange (Swedish Space Corporation sounding rocket launching range), near Kiruna, Sweden (68°N, 21°E) and Fairbanks, Alaska (65°N, 148°W).

Esrange, Kiruna

The Limb Profile Monitor of the Atmosphere (LPMA) is a solar occultation Fourier transform infrared (FTIR) spectrometer [*Camy-Peyret et al.*, 1993]. The measurements were made during the balloon ascent and at float (occultation at local sunset).

The Michelson Interferometer for Passive Atmospheric Sounding-Balloon-borne Version 2 (MIPAS-B2) is a cryogenic FTIR spectrometer that measures atmospheric thermal emissions from the limb [*Oelhaf et al.*, 1996; *Friedl-Vallon et al.*, 1999]. The measurements were made during the balloon float in the nighttime.

The Système d'Analyse par Observation Zénitale (SAOZ)-BAL is a balloon-borne UV-visible spectrometer [*Pommereau and Piquard*, 1994], which makes use of the solar occultation technique from the balloon. The measurements were made during the balloon ascent and at float (occultation at local sunset).

The Cold Atmospheric Emission Spectral Radiometer (CAESR) is a scanning monochromator that measures thermal emission from the atmosphere [*Murcray et al.*, 1994]. The measurements were made during the balloon ascent. It was launched together with the chemiluminescence detector (CLD) instrument on February 10 and 25, 1997, and with the MkIV instrument on May 8, 1997 from Fairbanks.

CLD is an in situ NO_y (NO + NO₂ + NO₃ + HNO₃ + $2(N_2O_5)$ + HO₂NO₂ + ClONO₂ + BrONO₂ + aerosol nitrate) sensor based on the technique using the NO/O₃ chemiluminescence detection after catalytic conversion of the component species into NO on a surface of gold tubes heated to 300°C [*Kondo et al.*, 1999]. The data were recorded every 5 s during both the ascent and parachute descent. The ascent and descent data agreed well, and 1 km average concentrations were used in this study. The amount of HNO₃ was estimated from the measured NO_y mixing ratio with HNO₃/NO_y ratios calculated using a box model developed by Atmospheric and Environment Research, Inc. (AER) [*Danilin et al.*, 1998], as described in *Koike et al.* [2000].

The ASTRID is an in situ grab sampler [*Bauer et al.*, 1994]. The measurements were made during the controlled slow descent of the balloon.

The BONBON [*Schmidt et al.*, 1991] and SAKURA [*Honda et al.*, 1996] are in situ cryogenic samplers. The measurements were made during the controlled slow descent of the balloons.

Besides these established balloon sensors, we also used HNO_3 (from NO_y) data measured with chemiluminescence NO_y detector aboard the Deutsches Zentrum für Luft-und Raumfahrt (DLR) Falcon research aircraft [see *Koike et al.*, 2000].

Fairbanks

The MkIV is a solar occultation FTIR spectrometer [*Toon*, 1991] that measures the entire 650-5650 cm⁻¹ spectral region simultaneously with a 0.01 cm⁻¹ resolution. The

measurements were made during the balloon float (38 km altitude) at sunrise.

The Far-Infrared Spectrometer (FIRS)-2 is a FTIR spectrometer that measures atmospheric thermal emission in the wavenumber range of 75-1300 cm⁻¹ [*Johnson et al.*, 1995]. The limb soundings were made when the balloon was at float altitude of 40 km.

3. Differences in the ILAS Version 5.20 and 6 retrieval algorithms

Detailed description of the Version 5.20 retrieval algorithm and the tangent height determination method was given by *Yokota et al.* [2002] and *Nakajima et al.* [2002b], respectively. Here we describe the differences between Version 6 and Version 5.20 algorithms. A statistical comparison of vertical profiles of each species obtained by the two Versions are shown in Appendix 2.

One of the main differences between the two retrieval strategies is the target gas selection. In the Version 5.20, O₃, HNO₃, NO₂, N₂O, CH₄, H₂O, CFC-11, CFC-12, and COF₂ were retrieved simultaneously. It has been found that we miscalculated absorption cross-sections for CFC-11, CFC-12, and N₂O₅ in the look-up table when using the pseudo-line data set provided by *G. C. Toon* [private communication, 1995]. (Note that the pseudo-line data themselves are correct.) The absorption cross-sections for CFC-11, CFC-12, and N₂O₅ were underestimated by about 60-70% in the Version 5.20. Correction for this has been made in Version 6, which generates N₂O₅ products reasonably good (but not validated quantitatively yet). Also, the quality in the CFC-12 data has been significantly improved [*Khosrawi et al.*, 2004]. Moreover, the pseudo-line data of ClONO₂ has been updated by *G. C. Toon* [private communication, 2003] on the basis of laboratory measurement [*Wagner and Birk*, 2003]. This has been introduced to the Version 6 retrieval algorithm, and ClONO₂ also has been revealed to have reasonable features in its data quality [*Nakajima et al.*, 2005]. In summary, ClONO₂ and N₂O₅ has been changed from the fixed gas in

the Version 6. Consequently, we need the climatological value of COF_2 profiles as one of the fixed gases. Using the ATMOS Version 3 data set [*Irion et al.*, 2002], they were calculated hemispherically and half-yearly, and were newly prepared for the Version 6 retrieval algorithm.

The other main points that have been revised or modified from the Version 5.20 algorithm are: (1) updated line parameters of the O_2 A band [*Rothman et al.*, 2003] for determination of the tangent height, a high-resolution solar spectrum newly obtained from a balloon measurement [*Camy-Peyret et al.*, 2001], re-determined instrument functions of the visible/near-IR spectrometer, (2) molecular spectroscopic parameters from the HITRAN 2000 data set, a new H₂O continuum data set in the LBLRTM, (3) climatological gas and temperature/pressure profiles for initial values of retrievals and for the non-gaseous component correction, and (4) a method for interpolating the level 1 data into the respective layer boundary altitudes and a method for calculating the optical thickness within the respective atmospheric layers.

In addition, revisions were made for data processing of consecutively-missing major frames in the atmospheric measurement part and 100%- or 0%-radiance measurement part. As a result, about 300 more scenes were successfully processed in the Version 6 than Version 5.2. Algorithm for determining the lowest detectable altitude has been also revised, in which a function similar to a logarithmic curve was applied to the rising (falling) edge of the time-series of measured signals, making the detectable altitude lower (down to 7 km at the lowest case).

3.1. Revised determination of the tangent height

ILAS has employed the hybrid method (Hybrid-M), which is a combination of the transmittance-spectrum method (TS-M) and the sun-edge sensor method (SES-M) for determining tangent heights since Version 5.20 [*Nakajima et al.*, 2002]. The Hybrid-M

assumes that the TS-M can determine correctly the tangent height at an altitude of 30 km. The modifications to the methods for determining the tangent height are described for each of TS-M, SES-M, and others related to tangent height.

3.1.1. Modifications to the TS-M

(1) Molecular spectroscopic data in the O₂ A-band

The database containing $O_2 A$ band line parameters has been changed from HITRAN 1996 to HITRAN 2000.

(2) Intralayer interpolation for the theoretical transmittance area

The interpolation method for determining the theoretical transmittance areas for individual major frames was changed from Linear to Spline. The theoretical transmittance area is calculated at each 1-km (integer) grid.

(3) The Doppler effect on theoretical transmittance calculation at 100% radiance value

The Doppler effect due to the satellite movement toward the sun, which was not taken into account in the previous Versions, has been introduced in calculating the theoretical transmittance at 100% radiance data in the visible channel.

(4) The Doppler effect on the O₃ Wulf band cross section

The Doppler effect due to the satellite movement toward the sun has also been introduced in applying the O_3 Wulf band cross section used for correcting the baseline in the visible channel data.

(5) Air-to-vacuum wavelength conversion applied to the O₃ Wulf band cross section

The vacuum wavelength converted from the air wavelength is now correctly used for the cross section data in the O_3 Wulf band.

(6) High-resolution solar spectrum

The solar line data used in calculating the theoretical transmittance in the visible channel has been changed from the data in MODTRAN to data observed by LPMA [*Camy-Peyret et*]

al., 2001].

(7) Re-calibration of instrument functions of the visible channel

The wavelength center for instrument functions has been re-calibrated using the solar line data observed by LPMA [*Camy-Peyret et al.*, 2001].

(8) Re-definition of the range of spectral elements

In view of the revisions of 3.1.1.(1) molecular spectroscopic parameters, 3.1.1.(6) solar spectrum, and 3.1.1.(7) instrument functions, the ranges of spectral elements used for computing the transmittance area that are less subject to any uncertainty in the UKMO temperature have been re-determined.

3.1.2. Modifications to the SES-M

(1) Solar position calculation routine

The solar position was calculated from the table in *Newcomb* [1895] using a vernal equinox of J1997.5 in the previous Versions. This calculation method has been changed to another one based on the "true of date" coordinates.

(2) Local radius of the earth

The method for calculating the local radius based on the daily mean latitude (computed from about 14 scenes) for each hemisphere has been changed to that based on the observed latitude at each scene.

(3) Determination and correction of spike noise inherent in the sun-edge sensor data

The median filter is applied in the time-series direction as was done in the past. To avoid the mistake that normal data is taken to be abnormal, the filter width has been decreased from 19 major frames to 5 major frames while the noise threshold has been increased from 20 to 60 counts.

3.1.3. Other revisions related to TH

(1) Time determination

To minimize errors of time determination, the time for each major frame is calculated once and then the regression line is drawn by using the times for all major frames (about 4500 data points) over one event. Accordingly, the time for each major frame has been redefined.

(2) Dead time t_0 in lock-in amplifier response for infrared channel

The dead time t_0 in the lock-in amplifier response for the infrared channel has been changed from 65 msec to 44 msec to take into account the delay (21 msec) in data acquisition timing among channels.

(3) Determination logic for minimum detectable altitude

Old: The rising (falling) edges of time-series signals were determined by means of the 2nd-order difference values, leading to erroneous results due to noise in some cases. **New:** A function like a logistic curve is applied to the rising (falling) edges of time-series signals to determine the major frame number that corresponds to the lowest altitude, as long as the stabilization of the function has been kept. In addition, for data around the endpoint of the time-series signals out of range of a digital filter, the data have been replaced by linear extrapolation for the time-series signals.

3.2. Modifications to molecular spectroscopic parameters

The updated molecular spectroscopic parameters and the revised handling of spectroscopic data are described below.

(1) PT table of cross section data for infrared channel

The HITRAN 1996-based PT table (a look-up table utilizing temperature and pressure data) has been changed to the HITRAN 2000-based PT table and the partition function has been updated. See Table 4 for more information.

(2) H₂O continuum

The LBLRTM H₂O continuum CKD has been changed from Version 2.1 to Version 2.4. (3) H₂O PT table

The H₂O continuum mentioned in 3.2.(2) above includes the absorptions for the portions in both wings 25 cm⁻¹ apart from the line center, as well as the absorption for a rectangular area whose height is equivalent to an absorption amount at ± 25 cm⁻¹ from the line center and whose width is 50 cm⁻¹, among the absorptions computed from the H₂O line parameters. **Old:** The H₂O PT table excluded the absorptions of both wings (area 1), but included the absorption for the rectangular area (area 2) mentioned above.

<u>New:</u> According to the definition above, the new H_2O PT table excludes both area 1 and 2 correctly.

(4) Interpolation of temperature-dependent cross section data of CF₄

<u>Old:</u> The nearest data was extracted from several temperature-dependent cross section data points surrounding the target point.

New: Linear interpolation was applied between the temperature-dependent cross section data points.

(5) Line parameters for $O_2 A$ band

See the description in 3.1.(1) above.

3.3. Modification for climatological values for gas, temperature, and pressure profiles

The modifications to the climatological (initial) values for gas and temperature/pressure profiles are described below.

(1) Climatological values for gas profiles

<u>Old</u>: Assigned the same values as those at the lowest altitude of climatological profiles below the lowest altitude.

<u>New:</u> For the following gases, their average concentrations in the troposphere in 1996-1997 are set below 9 km altitude (CFC11 = 270 pptv, CFC12 = 530 pptv, N₂O = 312 ppbv, CH₄ =

1730 ppbv). In addition, the mixing ratios of NO_2 , HNO_3 , and $ClONO_2$ are set to decrease with decreasing or increasing altitudes with a scale height of 3 km for the altitude range where no climatological values exist.

(2) Upper stratosphere/lower mesosphere data in the initial profile of temperature/pressure
 Old: Created from UKMO and CIRA86 (at an altitude of approx. 50 km or higher).
 New: Created from UKMO^{\$}, mean profiles from any of HALOE, MLS, GPS/MET[#], and CIRA86[&].

^{\$} Applied to altitudes below 40 km for each scene.

[#] Applied to altitudes from 50 km to 60 km. Created from the monthly and latitude-based mean profiles of the datasets which were collected during the ILAS observation period.

[&] Applied to altitudes above 65-70 km.

3.4. Other modifications

(1) Instrument functions of infrared channel

The results from laboratory experiments based on gas cells were reanalyzed, and the instrument functions were re-determined accordingly.

(2) Signals from infrared and visible channels interpolated into the layer boundary

<u>Old</u>: The nearest major frame to the layer boundary altitude of each 1-km (integer) grid was selected from the top of each layer.

New: Data are linearly interpolated into the layer boundary altitude from the two major frames interposing it.

(3) Intralayer distributions

The method for calculating the representative value of the optical thickness of the atmospheric layers was changed from the rough approximation method to a more precise approximation method.

(4) Processing of consecutively missing major frames in the atmospheric measurement part

<u>Old:</u> If any consecutively-missing major frames were found in the atmospheric measurement part, processing was aborted.

New: Linear interpolation is applied up to four consecutively-missing major frames for data correction.

(5) Processing of 100%- and 0%-radiance part

<u>Old:</u> If any consecutively-missing major frames were found in the 100%- and 0%-radiance part, processing was aborted.

New: If 80 or more major frames are found in each of those parts (and also at least 20 major frames found in both of the 0% parts before and after observation), processing is continued.

The modifications in 3.4.(4) and 3.4.(5) allow more than 300 scenes of data to be finally processed.

(6) Calculation of AEC at 780 nm

Old: Extinctions due to Rayleigh scattering were subtracted according to Frohlich and

Shaw [1980] with the assumption that the King correction factor is unity.

New: The experimental formula proposed by Chance and Spurr [1997] is applied.

As a result, about 5% of the Rayleigh extinction coefficient in Version 6 were reduced compared to those in Version 5.2 for all altitudes.

4. Comparisons with other sensors

4.1. Data selection

Criteria for comparisons used in this study are completely the same as those for the validation analyses so far conducted. Namely, see *Sugita et al.* [2002] for O_3 , *Koike et al.* [2000] and *Irie et al.* [2002] for NO_2 and HNO_3 , *Kanzawa et al.* [2003b] for N_2O and CH_4 , *Kanzawa et al.* [2002; 2003a] for H_2O , *Hayashida et al.* [2000] and *Saitoh et al.* [2002] for AEC at 780 nm.

Here, we used criteria in universal time (UTC) and space differences to be ± 12 hours (or

±2 hours) and 300 km, respectively, in order to extract the coincidence pairs between ILAS and HALOE (or SAGE II). For searching coincidence measurements, location and time at TH of 20 km were used as the representative location and time of each measurement. Since SAGE II and HALOE are carried on inclined-orbit satellites, the occultation events occur globally. Therefore, coincidence measurements of SAGE II and HALOE with ILAS were limited in time and space. With the criteria defined above, 85 coincidence pairs for SAGE II and ILAS (hereafter referred to as SAGE II:ILAS), 202 pairs for HALOE:ILAS were selected in a first step. A summary of the coincidence measurements of satellite sensors is listed in Table 5, separately for the periods shown in the table. Although we could find 52 measurement pairs in November 1996 in the SH for SAGE II:ILAS, we omit to show the result from that period, because most of the data were taken inside the polar vortex.

For measurements obtained with balloon-borne instruments, we just used the nearest ILAS measurements to them. As the representative location and time of these measurements, those at the measurement (or tangent) point of 20 km altitude were used if such information was available (otherwise the locations at each launch site were used).

4.2. Consistency of altitude to be compared

In order to compare profiles between ILAS and validation data, the vertical grid (or resolution) should be consistent with each other. THs from satellite sensors are registered as geometric altitude with different spacing. For the HALOE data, we first linearly interpolated partial pressures of O_3 , CH_4 , and H_2O and atmospheric pressures to 0.1 km altitude grid. Then these data were averaged within each 1 km altitude bin centered at each *i* km grid (*i*: integer) to generate mixing ratios of O_3 , CH_4 , and H_2O . For the SAGE II data, the AEC data were averaged every 3 points centered at *i* km.

For balloon-borne instruments with a vertical resolution comparable to or better than ILAS, we just compared them at every 1 km geometric height interval. Altitudes for

several balloon-borne instruments, such as CLD and DLR NO_y detector, are registered with a better resolution than ILAS. In these cases, partial pressures of gaseous species and atmospheric pressures were integrated within each 1 km bin centered at each *i* km grid to generate mixing ratios. For FIRS-2, which has a lower altitude resolution than ILAS, we also compared it at every 1 km grid. For air-sampling sensors, such as ASTRID, BONBON, and SAKURA, we interpolated the profiles to every 1 km grid. For this comparative study, we used data obtained between 7 and 70 km for comparisons with satellite-borne and balloon-borne instruments, within the available data range.

4.3. Results from satellite sensors

4.3.1. HALOE

The volume mixing ratio profiles of O₃, CH₄, and H₂O obtained with the Version 6 ILAS retrieval algorithm are compared with those obtained with the Version 19 HALOE retrieval algorithm. As discussed in section 4.1, 202 coincident measurement pairs were selected Considering the relative position of the two measurements with respect to the polar first. vortex, data points of the coincidence pairs selected by the above criteria were further screened by the following procedure, as described detail in Sugita et al. [2002]. PV values at each tangent height location and time for ILAS and for its coincidence measurements were calculated. PV values and potential temperatures were calculated at each grid point. These data were then interpolated in time and space to each measurement (both for ILAS and HALOE measurements) with a 1-km geometric altitude grid. We defined the PV relative percentage difference as 100 * 2 * [PV(ILAS) - PV(HALOE)]/[PV(ILAS) + PV(HALOE)] in percent (%) for each 1-km altitude grid where PV(ILAS) and PV(HALOE) denote PV values at the time and location of ILAS and its coincidence validation measurement. If the relative percentage difference exceeds $\pm 15\%$ at consecutive altitude grids for more than 3 km, the data at these altitudes were discarded from the validation Then, if the retrieved value was smaller than its measurement uncertainty (which analysis.

was described in section 2), it was also filtered out. This data filtering generally excluded comparisons of small retrieved values with large relative errors.

<u>O</u>₃

Figures 3a and 3b show average profiles of O_3 obtained by ILAS and HALOE between 9 and 70 km in the NH and SH, respectively, together with minimum, maximum, and 1 sigma standard deviation of the data (left panel). In the right hand panel, the median of individual percentage differences, D, between ILAS and HALOE is shown, together with its minimum and maximum. Here, D is a relative difference defined as:

$$D(\%) = 100 \times 2 \times [X(ILAS) - X(HALOE)]/[X(ILAS) + X(HALOE)] (1)$$

where X(ILAS) and X(HALOE) show mixing ratios of X (any of O₃, CH₄, and H₂O) measured by ILAS and HALOE at each geometric altitude grid, respectively. The number N of coincidences or measurement pairs at each altitude is shown on the right-hand side of the figure. Two dashed lines symmetrical with respect to the zero line show averages of RSS of the errors in the ILAS and HALOE measurements at each altitude, defined as:

RSS error (mean) =
$$\sum_{i}^{N} \sqrt{Err_{i}(ILAS)^{2} + Err_{i}(HALOE)^{2}} / N \quad (i = 1 \text{ to } N)$$
(2)

where Err(ILAS) and Err(HALOE), show total measurement errors of ILAS and HALOE data, respectively, which were described in section 2.

In the NH comparison (Figure 3a), the D values were within $\pm 10\%$ from 17 to 52 km and generally decreased with increasing altitude up to around 60 km at which the D values (10-18%) exceeded the RSS errors. In the SH comparison (Figure 3b), the D values were within $\pm 10\%$ between 15 and 59 km and reached -15% at 64 km at which the D values

corresponded to the RSS error. Below 14 km, the D values increased with decreasing altitude, reaching more than 40%.

In summary, no statistically significant bias in the Version 6 ILAS O_3 data was generally recognized, except for the altitude region around 60 km in the NH comparison. (We will discuss this discrepancy for this altitude range in Appendix 2, in terms of the temperature and pressure profiles used for the Version 6 retrievals.)

<u>CH</u>₄

Similarly to Figure 3, Figures 4a and 4b show average profiles of CH_4 obtained by ILAS and HALOE (left panel) and profiles of the D value and the RSS errors (right panel) between 16 and 60 km in the NH and SH, respectively. Here D is the relative percentage difference defined in equation (1) by setting "X" to "CH₄", and the RSS error is the total error defined in equation (2) for the CH_4 measurement of ILAS and HALOE.

In the NH comparison (Figure 4a), the D values were within $\pm 18\%$ from 16 to 58 km. In the SH comparison (Figure 4b), the D values were between -13 and 16% from 16 to 54 km.

<u>H₂O</u>

Similarly to Figure 1, Figures 5a and 5b show average profiles of H_2O obtained by ILAS and HALOE (left panel) and profiles of the D value and the RSS errors (right panel) between 9 and 70 km in the NH and SH, respectively. Again, D is the relative percentage difference defined in equation (1) by setting "X" to "H₂O", and the RSS error is the total error defined in equation (2) for the H₂O measurement of ILAS and HALOE.

In the NH comparison (Figure 5a), the D values were between -9 and 4% from 13 to 52 km, and decreased with increasing altitude up to 61 km at which the D value was -24%. In the SH comparison (Figure 5b), the D values were between -13 and 7% from 13 to 67 km.

4.3.2. SAGE II (AEC at 780 nm)

Figures 6a and 6b show profiles of D values between ILAS and SAGE II in May 1997 in the NH and in February 1997 in the SH, respectively. Here, D is also the relative percentage difference defined in equation (1) by setting "X" to "AEC" and HALOE replaced with SAGE II. RSS error is the total error defined in equation (2) for the AEC measurement of ILAS and SAGE II. The value of AEC at 780 nm for SAGE II data was calculated by linear interpolation in the double logarithmic space between wavelength and AECs at 525 nm and 1020 nm that were measured by SAGE II. In NH, D values ranged from -10 to -40%, exceeding the range of the RSS total error. However, at altitudes where the AEC values are high, D values ranged from zero to -20%. In SH, D values ranged from -20 to +5% below 18 km, except for the lowest altitude. Above 19 km, D values decrease with increasing altitude in both of the hemispheres. In these regions, the retrieved AEC values become so small that it generally difficult to make any comparisons.

4.4. Results from balloon-borne instruments (HNO₃, NO₂, and N₂O)

Figures 7 and 8 show comparisons between ILAS and its coincident balloon-borne sensors for HNO₃ and NO₂, respectively. The average of individual comparison between the two is shown in this figure. The difference is defined as absolute difference of each pair as ILAS - balloon in ppbv. For reference, comparisons between the former ILAS Version. 5.2 and the balloon sensors are also shown. The Version 6 HNO₃ data agree well with the balloon data within ± 0.5 ppbv, except for a point of 15 km altitude. The Version 6 NO₂ data also agree well with the balloon data within ± 0.5 ppbv, except for a point of 35 km altitude.

Figure 9 shows correlations between ILAS and its coincident balloon-borne sensors for N_2O . Data shown as black pluses indicate that the relative difference in PV values for each of the measured air masses at each altitude exceed 15%. Generally, ILAS and its

coincident balloon-borne sensors are well correlated each other, except for these black pluses. The root mean square difference from the one by one line is as small as 0.021 ppmv.

5. Concluding remarks

The Improved Limb Atmospheric Spectrometer (ILAS) aboard the Advanced Earth Observing Satellite (ADEOS) observed vertical profiles of ozone (O₃), nitric acid (HNO₃), nitrogen dioxide (NO₂), nitrous oxide (N₂O), methane (CH₄), water vapor (H₂O), and other several gaseous species such as chlorine nitrate (ClONO₂) or dichlorodifluoromethane (CF₂Cl₂, CFC-12), as well as aerosol extinction coefficient (AEC) at 780 nm in the high-latitude stratospheres from November 1996 through June 1997. The vertical profiles of O₃, HNO₃, NO₂, N₂O, CH₄, H₂O, and AEC at 780 nm retrieved by the Version 5.20 data processing algorithm were well validated [*Sasano*, 2002; and references therein]. In this report, these seven chemical species newly retrieved by the Version 6 algorithm were again compared with its correlative data from balloon-borne measurements and coincident data from satellite-borne measurements. The characteristics of Version 6 CFC-12 and ClONO₂ data are well evaluated elsewhere [*Khosrawi et al.*, 2004; *Nakajima et al.*, 2005].

The difference between the Version 5.20 and the Version 6 retrieval algorithm was described in detail and the quantitative difference between the two data set in each of the chemical species was also evaluated. For O₃, the differences, in terms of the percentage difference in (Version 6 - Version 5.2)/Version 5.2, range from -12% to -1% for altitudes between 11 and 50 km. For HNO₃, the differences range from -9% to +7% for altitudes between 16 and 40 km. For NO₂, the differences range from -13% to +13% for altitudes between 21 and 50 km. For N₂O, the differences range from -12% to 0% for altitudes between 11 and 30 km. For CH₄, the differences range from -11% to -2% for altitudes between 11 and 60 km. For H₂O, the differences range from -12% to +2% for altitudes

between 11 and 50 km. For AEC at 780 nm, the differences range from -17% to -12% for altitudes between 11 and 20 km.

The precision of the measurement was assessed by the minimum variability of the mixing ratio data. The quality of the Version 6 data set is generally comparable to that of the former Version 5.20 and available to the general public. The Version 6 ILAS data set includes more scenes (roughly 300) and covers lower detectable altitudes (down to 7 km) compared to the Version 5.20 data set. In total, the Version 6 data set has about 6100 measurement scenes.

Appendix 1: Updated error evaluations for the Version 6.1

It has been pointed out that the internal errors, which have been released as the Version 5.2 and 6 ILAS data products, may involve systematic errors inherent in the calculated and/or measured spectra. Basically, internal errors are estimated on the basis of the assumption that spectral residuals are due to random errors, and the spectral residuals are evenly provided for the respective gas species. Recently, however, an in-depth study on the spectral residuals has revealed that systematic spectral residuals of non-negligible magnitude exist in the respective measurement events obtained by ILAS in common. In principle, this type of systematic deviation of spectra represents the biases of retrieved gas concentrations, and so should not be considered as internal errors (random errors). As a result, the internal errors as estimated for the Version 6 (as well as for the Version 5.2) have caused significantly large relative errors, particularly for minor gas species.

With this in mind, error calculation method has been updated as the Version 6.1 data product. As for ILAS Version 6 product, the systematic structure is recognized in the residual spectra.

Internal error =
$$\left(diag \left(\frac{(\hat{\tau} - \tau)^t \cdot W \cdot (\hat{\tau} - \tau)}{44 - n} \right) \cdot (J^t \cdot W \cdot J)^{-1} \right)^{\frac{1}{2}}$$
 (A1)

If a residual sum of squares including systematic residuals (the part of diagonal of the matrix (i.e. diag()) in the right side of the equation(A1)) were used in computing internal errors, the errors would be over-estimated. (Since the diag() is a Scalar quantity, namely a constant value, the retrieval error for each gas increases at the same rate.)

An improved method is as follows. Systematic residuals are taken to come from unknown external error factors. Consequently, in the calculation of internal errors, components of estimated systematic residuals are subtracted from the squared sum of spectral residuals. On the other hand, the components of estimated systematic residuals are added to the term in the external errors.

[Methods of separation of systematic residuals from internal error bars]

As the first approximation for the systematic residuals, averaged values for each of the northern and the southern hemispheres, and for each tangent height, $ave(\Delta \tau_s(h))$, $ave(\Delta \tau_r(h))$ are calculated from the residuals after the retrieval of data for the whole period. The internal errors excluding the systematic residuals are calculated from the following equation (A2).

Internal error =
$$\left(diag\left(\frac{{}^{t}\left(\hat{\tau}-\tau-ave(\Delta\tau_{*}(h))\right)\cdot W\cdot(\hat{\tau}-\tau-ave(\Delta\tau_{*}(h)))}{44-n}\right)\cdot \left({}^{t}J\cdot W\cdot J\right)^{-1}\right)^{\frac{1}{2}}$$
(A2)

Then the estimated value, caused by the systematic residuals, would newly be added to the external errors resulted from the climatological data, as follows:

External error =
$$\sqrt{ext._{original}^{2} + ext._{unknown}^{2}}$$
 (A3)

Here, ext._{original}: The errors resulted from the climatological data used in the non-gaseous correction, (These are the same as the conventional External errors of Version 6), and Ext._{unknown}: The errors resulted from the unknown external factors, which are caused by estimated systematic residuals of the spectra, and were calculated in equation (A4) as follows:

$$ext_{unknown} = \left(diag\left({}^{t}J \cdot W \cdot J'\right)^{-1}\right)^{\frac{1}{2}}$$

$$W' = \left(\begin{vmatrix} \frac{1}{ave(\Delta \tau_{*}(h))_{1}} \end{vmatrix}^{2} \cdots 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & \left| \frac{1}{ave(\Delta \tau_{*}(h))_{n}} \right|^{2} \end{matrix}\right)$$
(A4)

J: Mean value of Jacobian for each of the northern and southern hemispheres, and for each tangent height:

It should be noted that the evaluation equation for $ext._{unknown}$ should be used only when the observed value for each detector element originally has a random error in the almost same order of $ave(\Delta \tau_*(h))$.

Systematic residuals of spectra, which are caused by the unknown factors, are rest of the fitted spectra which can not explained by the bias of the gas retrieved value from the true value (so called the "bias error of the gas retrieved values").

The component of the systematic residuals of spectral explained by the gas retrieval bias is actually unknown, however, here, we assume that the primary approximation may be almost the same amount of the estimated systematic residuals of spectra. Hence, we added it newly to the external error. The result of new internal and total error values are shown in Figures A1-1a and A1-1b, in the same manner as Figures 1a and 1b. Table A1-1 also shows the value of the error analyses. Note that the retrieved values are completely identical between the Version 6 and 6.1. The open public data set includes this new error estimate, and the Version 6.1 data set is released through http://www-ilas.nies.go.jp/.

Appendix 2: Comparisons between Version 5.2 and Version 6 profiles

Figures A2-1 to A2-8 show the mean profiles of gaseous chemical species in mixing ratio and AEC at 780 nm for all of the NH data. The data numbers are that both of Version 5.2 and Version 6 exist (2894 scenes). The profile shown in the right panel is calculated from the average of individual differences between the two Versions and the average of the Version 5.2. Results for all of the SH data are also shown in Figures A2-9 to A2-16.

Table A2-1 shows differences between the two Versions in terms of 100^* (Version 6 - Version 5.2)/Version 5.2 (%) for each of the species. The average of individual differences between the two Versions is divided by the average of Version 5.2 separately for hemispheres. The number weighted average for the respective 5 km altitude ranges are shown. In addition Table A2-2 shows differences between the two Versions of air number density which was calculated from the initial values of temperature and pressure (see Section 3.3.(2)). It should be noted that these differences (up to 9%) in the upper stratosphere directly compensate the differences found in chemical species; for instance, the difference in O₃ data in the NH for the 56-60 km altitude range (-15%) is explained by the difference in air number density of 9% at least. Figure A2-17 also shows correlations between the two Versions below 30 km altitudes for each of the chemical species.

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Figure captions

Figure 1a. Repeatability (cyan), internal error (blue), and total error (red) of the Version 6 data for each of the chemical species for the NH. See text (Section 2.2) for detail. Values are also shown in Table 1a.

Figure 1b. Same as Figure 1a, but in the SH.

Figure 2. Bias error caused by the nongaseous correction for a) O_3 , b) HNO₃, c) NO₂, d) N₂O, e) CH₄, f) H₂O, g) CFC12, and h) ClONO₂. Bias errors expressed in gas number density (cm-1) are presented as a function of the AEC at 780 nm for various aerosols and PSCs. The scales of volume mixing ratios (ppmv) for altitudes of 15, 20, and 25 km are also shown at the right-hand side of each chart. The component ratios from STS(a) to STS(d) are the same as described in the notes of Table 2.

Figure 3a. Average profiles of ozone mixing ratios retrieved by ILAS (Version 6) and HALOE (Version 19) in the Northern Hemisphere (left panel). The ILAS data are plotted with 0.2 km shift for clarity. Numbers of coincidence measurement pairs at each altitude are shown on the right-hand side of the figure. Error bars show one sigma standard deviation of the data at each altitude. Maximum and minimum values of the data are shown as a solid line (ILAS) and a dotted line (HALOE), respectively. The median profile of individual percentage differences, labeled as D (see text for the definition), between ILAS and HALOE ozone mixing ratios is also shown (right panel). Maximum and minimum values of the data are shown as dash-dotted lines. Dashed lines symmetrical with respect to the zero line show the average of individual root-sum-square total uncertainties, labeled as RSS err., in ILAS and HALOE measurements (see text).

Figure 3b. Same as Figure 3a, but in the Southern Hemisphere.

Figure 4a. Same as Figure 3a, but for H_2O .

Figure 4b. Same as Figure 3a, but for H₂O and in the Southern Hemisphere.

Figure 5a. Same as Figure 3a, but for CH₄.

Figure 5b. Same as Figure 3a, but for CH₄ and in the Southern Hemisphere.

Figure 6a. The median profile (solid line) of individual percentage differences, labeled as D (see text for the definition), between ILAS and SAGE II AEC at 780 nm in May in the Northern Hemisphere. Dashed lines symmetrical with respect to the zero line show the average of individual root-sum-square total uncertainties, labeled as RSS err., in ILAS and SAGE II measurements (see text).

Figure 6b. Same as Figure 6a, but in the Southern Hemisphere.

Figure 7. The average profile of individual absolute differences between ILAS Version 6 and balloon HNO_3 (red line with open circles) in the Northern Hemisphere. A black line with open square shows for ILAS Version 5.2 for reference.

Figure 8. Same as Figure 7, but for NO₂.

Figure 9 Correlation of ILAS Version 6 versus balloon N_2O in the Northern Hemisphere. The black-colored marks show the pairs whose relative PV percentage differences are larger than 15%. Root-mean-square difference of ILAS data against balloon data (excluding the black-colored pairs) is shown to be 0.016 ppmv.

Figure A1-1a. Same as Figure 1a, but for V6.1

Figure A1-1b. Same as Figure 1a, but for V6.1 and in S.H.

Figure A2-1. Average profiles of ozone mixing ratios retrieved by Version 5.2 (blue) and Version 6 (red) of ILAS for all of the NH data that both of the Versions exist. Error bars show one sigma standard deviation of the data at each altitude. Maximum and minimum values of the data are shown. The average of individual differences (Version 5.2 - Version 6) divided by the average of the Version 5.2 is shown in the right panel. One sigma standard deviation, maximum, and minimum values of the data are also shown.

- Figure A2-2. Same as Figure A2-1, but for HNO₃.
- Figure A2-3. Same as Figure A2-1, but for NO₂.
- Figure A2-4. Same as Figure A2-1, but for N_2O .
- Figure A2-5. Same as Figure A2-1, but for CH₄.
- Figure A2-6. Same as Figure A2-1, but for H_2O .
- Figure A2-7. Same as Figure A2-1, but for CFC12.
- Figure A2-8. Same as Figure A2-1, but for AEC at 780 nm.
- Figure A2-9. Same as Figure A2-1, but for the SH..
- Figure A2-10. Same as Figure A2-1, but for HNO₃ and for the SH.

- Figure A2-11. Same as Figure A2-1, but for NO₂ and for the SH.
- Figure A2-12. Same as Figure A2-1, but for N₂O and for the SH.
- Figure A2-13. Same as Figure A2-1, but for CH₄ and for the SH.
- Figure A2-14. Same as Figure A2-1, but for H₂O and for the SH.
- Figure A2-15. Same as Figure A2-1, but for CFC12 and for the SH.
- Figure A2-16. Same as Figure A2-1, but for AEC at 780 nm and for the SH.

Figure A2-17 Correlation of ILAS Version 6 versus ILAS Version 5.2 below 30 km altitudes for each of the chemical species for all of the scenes. a) O_3 , b) HNO₃, c) NO₂, d) N₂O, e) H₂O, f) CH₄, and g) CFC12.

Altitude [km]	Rep. [%]	Int. Error [%]	Tot. Error [%]
10	NaN	0 ₃ 46	47
15	11	8	9
20	4	5	7
30	4	2	4
35	1	2	2
40	1	2	2
45	2	2	2
55	3	4	4
60	4	5	6
65	7	10	10
70	16	HNO.	70
10	NaN	89	117
15	17	10	14
20	4	6	7
30	4	21	21
35	11	100	100
40	45	285	285
45	252	162 NO ₂	162
10	NaN	NaN	NaN
15	49	74	82
20	7	55	57
30	2	28	29
35	3	31	31
40	3	56	56
45	10	148	148
50		N ₂ O	250
10	NaN	36	40
15	5	12	14
20	4	16	16
30	10	69	69
35	14	170	170
40	20	220	220
50	165	278	278
		CH ₄	
10	NaN	18	21
20	2	10	12
25	9	15	17
30	6	29	30
35	7	69 74	69 74
40	11	60	60
50	22	64	64
55	32	80	80
60	58	H ₂ O	100
10	NaN	6	15
15	3	5	7
20	2	5	6
30	2	7	8
35	2	12	12
40	2	15	15
45	3	10	10
55	8	23	23
60	12	27	28
65	16	31	31
70	15	CFC12	17
10	NaN	29	30
15	6	25	27
20	6	52 84	54 93
30	31	188	199
35	75	354	354
10	XT. X T	CIONO ₂	XT_X7
10	NaN 51	NaN 56	NaN 65
20	21	110	116
25	12	59	62
30	7	91	92
40	109	603	604
		AEC780nm	
10	14	7	8
15	9 7	3	4
25	129	NaN	NaN

Altitude [km]	Rep. [%]	Int. Error [%]	Tot. Error [%]
10	7	46	47
15	9	8	10
20	4	5	7
25	6	3	4
30	5	2	3
33	1	2	2
40	1	23	23
50	2	4	4
55	3	5	5
60	5	7	7
65	7	12	12
70	14	9	56
		HNO ₃	100
10	13	82	108
15	13	10	1/
20	4 2	0 7	8
30	3	19	19
35	10	99	99
40	55	312	312
45	314	170	170
	1	NO ₂	
10	NaN	NaN	NaN
15	25	47	51
20	6	35	35
23	4	21	21
35	2	28	28
40	3	44	20 44
45	9	109	109
50	37	271	271
	1	N ₂ O	
10	3	45	49
15	5	12	14
20	4	18	19
25	4	25	25
30	4	51	51
35	24	1/9	180
40	24 63	200	289
50	168	205	205
		CH4	
10	1	21	23
15	3	10	13
20	2	14	17
25	2	16	18
30	3	26	27
35	0	00 84	00 84
40	10	83	83
50	22	95	95
55	38	119	119
60	77	119	119
	1	H ₂ O	
10	1	7	18
15	2	5	7
20	2	5	7
25	1	6	7
30	1	12	8
55 40	2	12	12
45	3	15	17
50	5	22	22
55	8	29	29
60	13	34	34
65	16	38	38
70	14	17	18
10		CFC12	20
10	2	38	38
20	0	24	27 61
20	11	75	83
30	28	222	234
35	91	388	388
		CIONO ₂	
10	31	501	652
15	76	NaN	NaN
20	10	118	125
25	7	84	86
30	7	132	133
35 40	16	339 576	339 576
40	/3	AEC780nm	576
10	8	6	7
15	3	3	4
20	5	5	6
25	15	14	14

	а	1σ s.d. of	a (cm ⁻³)	15km(ppbv)	20km(ppbv)	25km(ppbv)
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	1. 43E+15 1. 61E+14 5. 96E+12 2. 64E+14 2. 79E+14 2. 37E+14 -9. 20E+13 2. 23E+14	4. 02E+13 1. 07E+13 1. 27E+12 1. 11E+13 1. 13E+13 8. 58E+12 1. 26E+11 1. 40E+12	3 + 12 1.43E+12 1.61E+11 5.96E+09 2.64E+11 2.79E+11 2.37E+11 -4.60E+10 1.11E+11	$\begin{array}{c} 392.\ 007\\ 44.\ 184\\ 1.\ 633\\ 72.\ 306\\ 76.\ 469\\ 65.\ 000\\ -12.\ 608\\ 30.\ 533\end{array}$	$\begin{array}{c} 856.\ 781\\ 96.\ 571\\ 3.\ 570\\ 158.\ 035\\ 167.\ 133\\ 142.\ 066\\ -27.\ 556\\ 66.\ 734 \end{array}$	$\begin{array}{c} 1897.\ 646\\ 213.\ 890\\ 7.\ 907\\ 350.\ 024\\ 370.\ 175\\ 314.\ 656\\ -61.\ 032\\ 147.\ 807 \end{array}$
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	-2. 81E+11 -2. 02E+12 -4. 38E+11 3. 49E+11 5. 00E+11 3. 06E+11 9. 46E+10 1. 45E+11	6. 38E+09 4. 43E+10 8. 41E+09 9. 88E+09 1. 31E+10 8. 01E+09 5. 21E+08 5. 79E+08	NO ₂ -2. 81E+08 -2. 02E+09 -4. 38E+08 3. 49E+08 5. 00E+08 3. 06E+08 4. 73E+07 7. 25E+07	$\begin{array}{c} -0.\ 077\\ -0.\ 554\\ -0.\ 120\\ 0.\ 096\\ 0.\ 137\\ 0.\ 084\\ 0.\ 013\\ 0.\ 020\\ \end{array}$	$\begin{array}{c} -0.\ 168\\ -1.\ 211\\ -0.\ 262\\ 0.\ 209\\ 0.\ 300\\ 0.\ 183\\ 0.\ 028\\ 0.\ 043 \end{array}$	$\begin{array}{c} -0.\ 373\\ -2.\ 683\\ -0.\ 581\\ 0.\ 463\\ 0.\ 664\\ 0.\ 405\\ 0.\ 063\\ 0.\ 096\end{array}$
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	3. 63E+11 -1. 21E+12 -3. 33E+09 1. 01E+11 1. 82E+11 2. 46E+11 3. 39E+11 1. 25E+11	2.77E+10 3.00E+10 8.31E+09 1.45E+09 4.20E+09 7.29E+09 1.93E+09 1.36E+09	$\begin{array}{c} 11NO_3\\ 3.\ 63E+08\\ -1.\ 21E+09\\ -3.\ 33E+06\\ 1.\ 01E+08\\ 1.\ 82E+08\\ 2.\ 46E+08\\ 1.\ 69E+08\\ 6.\ 24E+07\\ NO\end{array}$	$\begin{array}{c} 0.\ 099\\ -0.\ 333\\ -0.\ 001\\ 0.\ 028\\ 0.\ 050\\ 0.\ 067\\ 0.\ 046\\ 0.\ 017 \end{array}$	$\begin{array}{c} 0.\ 217\\ -0.\ 727\\ -0.\ 002\\ 0.\ 060\\ 0.\ 109\\ 0.\ 147\\ 0.\ 101\\ 0.\ 037\\ \end{array}$	$\begin{array}{c} 0.\ 482 \\ -1.\ 611 \\ -0.\ 004 \\ 0.\ 133 \\ 0.\ 241 \\ 0.\ 326 \\ 0.\ 225 \\ 0.\ 083 \end{array}$
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	2. 30E+12 -8. 45E+13 -1. 79E+13 3. 34E+12 7. 12E+12 2. 85E+12 1. 83E+13 1. 51E+12	6. 68E+11 4. 19E+12 9. 07E+11 1. 51E+12 2. 24E+12 2. 34E+12 1. 48E+12 6. 23E+11	1.20 2.30E+09 2.30E+09 2.45E+10 1.79E+10 3.34E+09 7.12E+09 2.85E+09 9.16E+09 7.53E+08 CU	$\begin{array}{c} 0.\ 630 \\ -23.\ 160 \\ -4.\ 914 \\ 0.\ 914 \\ 1.\ 952 \\ 0.\ 781 \\ 2.\ 510 \\ 0.\ 206 \end{array}$	$\begin{array}{c} 1.\ 378\\ -50.\ 620\\ -10.\ 741\\ 1.\ 999\\ 4.\ 266\\ 1.\ 708\\ 5.\ 487\\ 0.\ 451\end{array}$	$\begin{array}{c} 3.\ 051 \\ -112.\ 116 \\ -23.\ 789 \\ 4.\ 427 \\ 9.\ 448 \\ 3.\ 783 \\ 12.\ 153 \\ 0.\ 999 \end{array}$
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	-8. 94E+13 8. 08E+14 2. 95E+14 -1. 23E+14 -2. 16E+14 -1. 46E+14 -2. 96E+14 -1. 87E+14	3. 88E+12 5. 05E+13 6. 77E+12 5. 33E+12 7. 01E+12 2. 92E+12 3. 79E+12 3. 38E+12	-8. 94E+10 8. 08E+11 2. 95E+11 -1. 23E+11 -2. 16E+11 -1. 46E+11 -1. 48E+11 -9. 33E+10	$\begin{array}{c} -24.\ 497\\ 221.\ 278\\ 80.\ 712\\ -33.\ 792\\ -59.\ 140\\ -40.\ 120\\ -40.\ 496\\ -25.\ 574\end{array}$	$\begin{array}{r} -53.\ 541\\ 483.\ 630\\ 176.\ 407\\ -73.\ 858\\ -129.\ 259\\ -87.\ 687\\ -88.\ 510\\ -55.\ 894\end{array}$	-118.586 1071.171 390.716 -163.584 -286.290 -194.214 -196.036 -123.798
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	$\begin{array}{c} -3.\ 03E+14\\ -4.\ 74E+15\\ -1.\ 15E+15\\ 1.\ 16E+13\\ 1.\ 10E+14\\ -1.\ 22E+14\\ -1.\ 62E+14\\ -1.\ 28E+14 \end{array}$	9. 30E+12 1. 08E+14 3. 76E+13 8. 18E+11 5. 39E+12 1. 24E+12 9. 08E+11 8. 14E+11	n_20 -3. 03E+11 -4. 74E+12 -1. 15E+12 1. 16E+10 1. 10E+11 -1. 22E+11 -8. 08E+10 -6. 42E+10	$\begin{array}{r} -83.\ 022\\ -1297.\ 629\\ -315.\ 248\\ 3.\ 175\\ 30.\ 220\\ -33.\ 524\\ -22.\ 134\\ -17.\ 587\end{array}$	$\begin{array}{c} -181.\ 455\\ -2836.\ 135\\ -689.\ 015\\ 6.\ 938\\ 66.\ 050\\ -73.\ 271\\ -48.\ 377\\ -38.\ 438\end{array}$	$\begin{array}{r} -401.\ 896\\ -6281.\ 626\\ -1526.\ 068\\ 15.\ 368\\ 146.\ 292\\ -162.\ 284\\ -107.\ 147\\ -85.\ 135\end{array}$
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	3.37E+11 1.10E+11 -2.70E+09 9.10E+10 1.11E+11 1.18E+11 9.98E+10 7.56E+10	3. 45E+10 1. 12E+10 1. 02E+09 1. 09E+10 1. 12E+10 9. 18E+09 6. 68E+08 1. 45E+09	CFC12 3. 37E+08 1. 10E+08 -2. 70E+06 9. 10E+07 1. 11E+08 1. 18E+08 4. 99E+07 3. 78E+07 C10V0	$\begin{array}{c} 0.\ 092\\ 0.\ 030\\ -0.\ 001\\ 0.\ 025\\ 0.\ 030\\ 0.\ 032\\ 0.\ 014\\ 0.\ 010\\ \end{array}$	$\begin{array}{c} 0.\ 202\\ 0.\ 066\\ -0.\ 002\\ 0.\ 054\\ 0.\ 066\\ 0.\ 071\\ 0.\ 030\\ 0.\ 023 \end{array}$	$\begin{array}{c} 0.\ 447\\ 0.\ 145\\ -0.\ 004\\ 0.\ 121\\ 0.\ 147\\ 0.\ 157\\ 0.\ 066\\ 0.\ 050\\ \end{array}$
ICE NAT STS (5, 37) STS (33, 15) STS (47, 3) STS (60, 0. 5) S (75) S (50)	-7.07E+10 -1.99E+11 2.86E+10 -1.23E+11 -2.17E+11 -2.54E+11 -2.96E+11 -1.77E+11	3. 85E+09 2. 30E+10 7. 44E+09 3. 02E+10 4. 74E+10 5. 17E+10 2. 71E+10 1. 46E+10	-7. 07E+07 -1. 99E+08 2. 86E+07 -1. 23E+08 -2. 17E+08 -2. 54E+08 -1. 48E+08 -8. 83E+07	$\begin{array}{c} -0.\ 019\\ -0.\ 054\\ 0.\ 008\\ -0.\ 034\\ -0.\ 059\\ -0.\ 070\\ -0.\ 041\\ -0.\ 024 \end{array}$	$\begin{array}{c} -0.\ 042\\ -0.\ 119\\ 0.\ 017\\ -0.\ 073\\ -0.\ 130\\ -0.\ 152\\ -0.\ 089\\ -0.\ 053\end{array}$	$\begin{array}{c} -0.\ 094\\ -0.\ 264\\ 0.\ 038\\ -0.\ 163\\ -0.\ 287\\ -0.\ 337\\ -0.\ 196\\ -0.\ 117\end{array}$

Table 3.A list of spectroscopic parameters used for the version 5.2 and 6.

Ver.5.20 Ver.6	H96 ^b (line) H00 ^c (line), Dec. 2000, Apr. 2001	H96 (line) same as in the left	H96 (line) same as in the left	H96 (line), Dec. 2000	H96 (line) H00 (line), Feb. 2001	H96 (line) H00 (line), Sep. 2001	H96 (line), Dec. 2000	H96 (line) same as in the left	H96 (cross-section) Toon ^d (pseudo line), Nov. 2000	Toon (pseudo line) 1995 Toon (pseudo line), 2001	Toon(pseudo line) 1995 same as in the left	Toon(pseudo line) 1995 same as in the left	H96 (cross-section) H00 (cross-section),	Dec. 2000, Jun. 2001	LBLRTM CKD ^e Ver.2.1 LBLRTM CKD Ver.2.4	(continuum) (continuum)	
Ver.5.20	H96 ^b (line)	H96 (line)	H96 (line)	H96 (line)	H96 (line)	H96 (line)	H96 (line)	H96 (line)	H96 (cross-sec	Toon (pseudo	Toon(pseudo l	Toon(pseudo l	H96 (cross-sec		LBLRTM CKI	(continuum)	
Species	H_2O	CO_2	O ₃	N_2O	CH_4	NO_2	HNO ₃	COF_2	CIONO ₂	CFC-11	CFC-12	N_2O_5	CF_4		H_2O		
\mathbb{D}^{a}	-	5	3	4	9	10	12	29	35								

^a ID in the HITRAN database

^b The HITRAN 1996 database

^c The HITRAN 2000 database. The updated time is also shown.

^d Pseudo line data are provided by Dr. Geoffrey C. Toon, JPL/NASA

^e Sub-routine of Clough-Kneizys-Davies

Table 4.	A summary of correlative balloon-borne measurements.	Measurement location, time, accuracy, and vertical resolution for a TH of 20 km
are describ	bed, if such parameters are available.	

ice	94]	[6([6t	3]	94]	[9	14]	[]	[9]	5]	[[t	
Referen	ommereau and Piquard [199	Feigl et al. [199	Kondo et al. [199	Camy-Peyret et al. [199	Murcray et al. [199	Oelhaf et al. [199	Bauer et al. [199	Schmidt et al. [199	Honda et al. [199	Johnson et al. [199	Toon [199	
Vertical resolution (km)	1 F	ı	0.2-0.3	2.5	1.5	< 3	1	1	0	4	2	
Species ^c	NO2	HNO ₃	HNO ₃	NO ₂ , HNO ₃ , N ₂ O	HNO ₃	NO ₂ , HNO ₃ , N ₂ O	N_2O	N_2O	N_2O	HNO ₃ , N ₂ O	NO ₂ , HNO ₃ , N ₂ O	
Instrument	SAOZ	NOy detector ^d	CLD	LPMA	CAESR	MIPAS-B2	ASTRID	BONBON	SAKURA	FIRS-2	MkIV	
Id	JP. Pommereau and F. Goutail	H. Schlager	Y. Kondo	C. Camy-Peyret	F.J. Murcray	H. Oelhaf	A. Engel	A. Engel	T. Nakazawa and S.Aoki	W.A. Traub	G.C. Toon	
ðUT (hour) ^b	1.8 0.9	5.8 0.4	5.8 5.6	1.5 1.0	5.8 5.6 6.4	3.6	2.0 3.7	2.8	2.7 6.4	13.1	6.4	
Distance (km)	1010 620	310 380	437 262	853 616	437 262 740	200	348 110	341	136 758	640	740	
ngitude (deg) I	19.0 17.6	15.4 20.0	21.1 21.1	22.8 20.5	21.1 21.1 -146.3	30.1	24.4 26.0	26.5	27.3 28.6	-148.9	-146.3	
atitude (deg) Lc	67.4 66.9	68.9 69.7	67.9 67.9	65.8 66.9	67.9 67.9 68.6	9.69	68.0 68.5	68.0	68.1 65.6	69.3	68.6	
launch site La	Kiruna Kiruna	Kiruna Kiruna	Kiruna Kiruna	Kiruna Kiruna	Kiruna Kiruna Fairbanks	Kiruna	Kiruna Kiruna	Kiruna	Kiruna Kiruna	Fairbanks	Fairbanks	e format
Time (UT)	15:24 17:00	08:20 13:36	$09:10 \\ 09:18$	$14:40 \\ 15:30$	09:10 09:18 12:11	19:42	11:18 11:13	11:45	$11.53 \\ 10.45$	19:12	12:11	DD in the date
Date ^a	$970224 \\ 970320$	970128 970130	970210 970225	970214 970226	970210 970225 970508	970324	970210 970225	970211	970222 970318	970430	970508	^a YYMM

^b 1 structure for the compared between ILAS and other sensor ^c Species that used for this study ^d Air-borne instrument

Period	$No.^{a}$	Distance (km) ^b	Time (hr) ^c	Hemisphere ^d	Occultatio	n Mode ^e
					ILAS	Val.
HALOE:ILAS (To	otal No.: 202)					
Nov. 19 - 24	40	172 (28)	1.8 (3.5, 0.0)	HS	SS	SR
Dec. 10 - 16	39	204 (130)	1.6 (3.5, 0.0)	HS	SS	SS
Jan. 28 - 31	17	178 (19)	4.5 (5.3, 3.8)	HS	SS	SR
Feb. 18 - 20	22	165 (64)	0.5 (1.0, 0.1)	HS	SS	SS
Mar. 24 - Apr. 2	59	166 (23)	0.2 (0.5, 0.0)	HN	SR	SS
May 13 - 14	12	147 (9)	6.9 (7.4, 6.5)	HN	SR	SR
Jun. 16 - 18	13	248 (197)	$0.2 \ (0.4, \ 0.0)$	HN	SR	SS
SAGE II:ILAS (T	otal No.: 85)					
Feb. 3 - 4	20	196	0.5 (1.2, 0.0)	HS	SS	SS
May 28 - 30	13	229	$0.2 \ (0.4, \ 0.0)$	HN	SR	SS

A summary of coincident measurements for each period from November 1996 to June 1997. Table 5.

^a Numbers of coincident measurement profiles for each period.

^b Average of individual distance between observed locations with a criterion of 300 km separation. Minimum distance is shown in the parenthesis.

^c Average of individual time difference between observation times with a criterion of ± 12 hours (± 2 hours for SAGE II). Maximum and minimum time differences are shown in the parenthesis, respectively.

^d SH and NH are for solar occultations occurring in the Southern Hemisphere and the Northern Hemisphere, respectively.

^e SS and SR are for solar occultations occurring at sunset and sunrise as seen from the satellites, respectively.

Altitude [km]	Rep. [%]	Int. Error [%]	Tot. Error [%]
10	NaN	9	16
15	11	4	7
20	4	2	3
30	3	1	2
35	1	1	1
40 45	1	1	1
50	2	2	3
55	3	3	4
60	4	5	5
70	16	13	70
	HNO	3	
10	NaN	18	79
20	4	4	4
25	6	3	4
30	4	8	10
35 40	45	37	39 140
45	252	116	117
	NO ₂		
10	NaN 49	NaN 42	NaN 59
20	7	22	44
25	6	12	21
30	2	8	14
55 40	3	27	15
45	10	108	116
50	44	203	215
10	N ₂ O	8	20
15	5	6	12
20	4	6	7
25	11	10	12
30 35	10	26 62	50 64
40	20	101	105
45	45	180	182
50	165 CH.	1/3	173
10	NaN	4	12
15	2	5	9
20	2	4	10
30	6	12	15
35	7	25	26
40	9	36	3/
50	22	53	53
55	32	74	74
60	58 H.O	95	95
10	NaN	1	15
15	3	2	6
20	2	2	5
25 30	2	23	5 4
35	2	4	5
40	2	7	7
45 50	3	11	11
55	8	22	22
60	12	27	27
65 70	16	30	31
70	CFC	12	17
10	NaN	7	10
15	6	11	21
20	15	35	56
30	31	78	107
35	75	127	135
10	NaN	NaN	NaN
15	51	35	56
20	21	41	58
25 30	12	24	31
35	20	137	140
40	109	289	295
10	AEC 14	/80nm 7	Q
15	9	3	4
20	7	5 NoN	6 NoN
43	129	inain	INDIN

Altitude [km]	Rep. [%]	Int. Error [%]	Tot. Error [%]
10	7	10	14
15	9	4	8
20 25	4 6	2	6 4
30	3	1	2
35	1	1	1
40 45	1	1	2
50	2	3	3
55	3	5	5
60	5	11	12
70	14	9	56
10	HNO 13	3	71
15	13	5	15
20	4	2	4
25 30	23	3	4
35	10	39	44
40	55	157	176
45	314 NO ₂	109	113
10	NaN	NaN	NaN
15	25	25	35
20	4	17	28 18
30	2	9	17
35	2	11	17
40	9	24 75	82
50	37	227	241
10	N ₂ O	10	22
10	5	6	10
20	4	7	11
25	4	11	13
35	11	70	73
40	24	142	151
45 50	63 168	225	237
50	CH ₄	10)	110
10	1	5	16
20	3 2	5	12
25	2	8	10
30	3	12	14
40	10	44	46
45	13	56	59
50	22	81	84 114
60	77	118	120
10	H ₂ O	2	17
10	2	2	6
20	2	2	5
25	1	3	6
30	2	4 5	6
40	2	8	8
45	3	12	13
55	8	27	29
60	13	33	35
65 70	16	37	39 18
	CFC	12	
10	2	8	11
20	7	24	41
25	11	36	56
30	28	104	132
55	CION	149 1O ₂	100
10	31	146	484
15 20	76 10	NaN 52	NaN 78
20	7	43	52
30	7	66	76
35 40	16 73	136 307	142
-10	AEC	780nm	520
10	8	6	7
15	3 5	3	4
25	15	14	14

ILAS V06.00, Relative Internal & Total Error Northern Hemisphere V06.00 Repeatability V06.00 IntErr V06.00 TotErr 70 F Altitude [km] Altitude [km] Altitude [km] 40 60 Relative Error [%] 40 60 Relative Error [%] 40 60 Relative Error [%] CH4 H2O N2O Altitude [km] Altitude [km] Altitude [km] Relative Error [%] Relative Error [%] Relative Error [%] CIONO2 CFC12 AEC_780nm Repeatability Internal error **Total error** Altitude [km] Altitude [km] Altitude [km]

Relative Error [%]

Relative Error [%]

Relative Error [%]



Relative Error [%]

Relative Error [%]

Relative Error [%]



Figure 2





















ILAS V06.10, Relative Internal & Total Error Northern Hemisphere $_{O3}^{O3}$ V06.00 Repeatability V06.10 IntErr V06.10 TotErr 70 F Altitude [km] 05 Altitude [km] 05 Altitude [km] 40 60 Relative Error [%] 40 60 Relative Error [%] 40 60 Relative Error [%] CH4 H2O N2O Altitude [km] Altitude [km] Altitude [km] Relative Error [%] Relative Error [%] Relative Error [%] CFC12 CIONO2 AEC_780nm Repeatability Internal error **Total error** Altitude [km] Altitude [km] Altitude [km]

Relative Error [%]

Relative Error [%]

Relative Error [%]



Relative Error [%]

Relative Error [%]

Relative Error [%]



Figure A2-2



Figure A2-3



Figure A2-4



Figure A2-5





Figure A2-7



Figure A2-8




Figure A2-10



Figure A2-11



Figure A2-12



Figure A2-13



Figure A2-14



Figure A2-15



Figure A2-16









