

**A study on depletion of the upper stratospheric ozone in the
Antarctic from Umkehr ozone profile**

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Long-term measurements of the total ozone by ground-based and satellite instruments show a large decrease from the 1980s to the middle of 1990s over mid-high latitude. A record-breaking depletion of the ozone layer over the Antarctic region was observed during the springtime in 2006. The principal causes of ozone destruction are attributed to the ozone-depleting substances (ODS) including chlorofluorocarbon compounds (CFCs). Inside the polar vortex the ozone destruction rate is accelerated through complicated heterogeneous chemical reactions under the atmospheric condition with extremely low stratospheric temperature. Ozone observation in the upper stratosphere has been globally (about 60N-60S latitude) made by various satellites (SAGE: Stratospheric Aerosol and Gas Experiment, SBUV: Solar Backscatter Ultraviolet, HALOE: Halogen Occultation Experiment) since 1979. The long-term trend of ozone decrease is enhanced at higher latitude, and that decrease is larger in the Southern Hemisphere than the Northern Hemisphere.

The Dobson ozone spectrophotometer is the primary ground-based instrument making measurements of stratospheric ozone. The Umkehr observations made prior to the satellite measurements gave highly valuable information on the past vertical distribution of ozone in the atmosphere that guided the current understanding of changes in stratospheric ozone. The Umkehr observations made now provide a baseline to augment the various satellite measurements. However, the problem of the shift error related to replacement of the instrument at a station is known from the long-term Umkehr N-value (intensity ratio) measurement record archived in the World Ozone and Ultraviolet Data Centre (WOUDC) in Toronto, Canada. With regards to the issue of uncertainty of the long-term Umkehr ozone profile dataset, we investigate various measurement errors such as the N-value shift, and discuss a long-term record of ozone depletion over Antarctica. Multiple shifts in the Japanese network Umkehr data record have been associated with instrument replacements. Therefore, N-value data were reevaluated based on instrument intercomparisons. The newest UMK04 (Umkehr Retrieval Algorithm 2004) ozone profile retrieval algorithm is applied in the processing of all reevaluated N-value time-series. The UMK04 provides a profile Averaging Kernel that is applied to other independent observational data for vertical smoothing, and helps to minimize vertical-resolution related differences in data comparisons. The reprocessed Umkehr profile dataset is analyzed for the long-term trend change in troposphere and through the entire stratosphere. It is found that the upper stratospheric ozone level over the Antarctic station Syowa (69.0S, 39.6E) stayed consistently low since 1990s. Extremely low values can be seen in the record over the last few years. On the other hand the international automation observation system was developed, which is dedicated to the long-term observation, while making reliable measurements at many atmospheric conditions with high quality of operational Umkehr data acquisition in the springtime. The outline of the paper in each chapter is shown as the following.

In Chapter 1, the issue of ambiguity in the Umkehr calibration process that affects retrieved ozone vertical profiles is investigated for each station. Although well-maintained Umkehr data

record is considered a valuable source of information of long-term changes in the ozone vertical profile, the Umkehr record at Japanese stations has obvious shifts. Majority of the shifts are related to the exchange of instruments for calibration (for total ozone measurements) and the replacement of Shimadzu instruments by Dobson ozone spectrophotometers. N-value data were recently reevaluated based on the results of instrument intercomparisons. The data analysis revealed systematic errors that depend on solar zenith angle, total ozone, and other instrumental factors. The quality and error of ozone profile retrieved with the newest algorithm (UMK04) is investigated. The difference in the results depends on the deployed retrieval algorithm (UMK92 vs. UMK04), where UMK04 uses non-varying monthly averaged ozone profile for a priori, while the UMK92 algorithm uses the a priori ozone profile that has total ozone dependency. The Umkehr Averaging Kernel (AK) is applied to other independent observational data to vertically smooth highly resolved ozone profiles such as available from balloon borne instruments (Ozonesonde soundings) or lidar measurements. The results show that the revised Umkehr ozone profiles show improved consistency with both types of ozone observations as compared to the old datasets, especially with regards to ozonesonde observations (difference of less than 5%). The reprocessed Umkehr profile data set is used for trend analysis. This analysis shows decrease of upper stratospheric ozone layer derived from the Antarctic Umkehr observation taken over Syowa station for the last 30 years, since 1977.

In Chapter 2, we present vertical ozone profiles from Dobson Umkehr measurements conducted at Syowa station in Antarctica since 1977. Introduction of highly automated measuring system to the Dobson instrument in 1994 at Syowa resulted in high quality data acquisition of Umkehr measurements. Short Umkehr measurement (A, C, and D pairs) has been routinely conducted at Syowa, with the exception of the polar summer when solar zenith angle is too small. In this study we discuss features of reevaluated N-value record at Syowa and the UMK04 retrieved ozone profiles. In the ozone record analysis, seasonal variation, effects of solar activity, QBO and aerosol, etc. signals are removed from the data. From 1977 to 2007, the springtime ozone values in layers 8 and 9 showed decrease of 9.6%/decade, and the layer 4 ozone shows decrease at 16.6%/decade. The ozone hole in 2006 developed to the largest size ever observed, and at Syowa total ozone of 114 DU was recorded. This is the lowest total ozone value since the beginning of observation in 1977. The Umkehr measurements also show extremely low ozone amount in all layers above Syowa. Especially, the record low ozone (close to complete ozone destruction) is found in layer 4 on September 27, 2006 and in layer 3 on October 15.

In Chapter 3, we discuss a long-term ozone trend determined from the newly re-processed Umkehr ozone profiles at Japanese stations. The long-term trend in upper stratospheric ozone from 1977 to 2008 is assessed, whereas the seasonal variation, effects of solar activity, QBO and aerosol, etc. are accounted for. Long-term variations of UMK04 retrieved ozone in the combined layer 8+ (8, 9 and 10) are shown for Sapporo, Tsukuba, Naha and Syowa stations. Linear trends for two time periods, prior to 1996 (from 1970 or 1977 till 1996) and after (1996-2008), are also shown for each station. Trend analyses suggest a significant decrease in the upper stratosphere over Japan during the 1980s. The upper stratospheric ozone levels at Tsukuba Station have shown a steady increase at 5%/decade rate after 1996. At the same time, a 7.7%/decade decrease in ozone is found in Umkehr

data taken at Sapporo Station, which indicates an even stronger ozone depleting rate as compared to ozone depletion rates prior to 1996. Over the Antarctic Syowa station, upper stratospheric ozone has been at the lowest level since 1990s. Especially low values can be seen in the last few years. Observed difference in the upper stratospheric ozone changes among several stations may be exhibiting the latitude dependence of ozone depletion. In order to assess the linear long-term trend in ozone over the Antarctic Syowa station, we conducted analysis of data collected during the first half (from 1977 to 1996) and the second half (from 1996 to 2008) of the record, and also separated data in austral springtime (from September to November) and summer (from December to March) seasons. The years 1988 and 2002 that had episodes of large-scale stratospheric sudden warming were removed prior to analysis. The result of the upper stratosphere analysis is the following. The long-term trend of annual average (springtime and summer), during the first half is $-7.9\%/decade$, while during the second half the Syowa record shows decrease at -5.2% . When the long-term trends of stratospheric ozone are compared between the springtime and summer seasons, the decrease in the first half of the record is found to be slightly larger in the summer than in the springtime, while in the second half of the record the springtime trend decreases even further, whereas the summer trend is leveled out. As one of the characteristics of the Syowa record, a linear trend tends to flatten out in the 1996-2005 time period after a significant decrease in 1977-1996, although recent data show a decreasing trend again. The result of lower stratosphere is the following. The long-term trend in the annual averaged ozone data appears to decrease rapidly in the first half ($-31.9\%/decade$) of the record, while it shows much smaller decrease in the second half ($-5.1\%/decade$). The seasonally separated long-term trends show the strongest decrease in low stratospheric ozone in the springtime of the first half ($-35.3\%/decade$) and still significant decrease in the second half ($-17.5\%/decade$), whereas the summer time ozone does not show any long-term changes in both the first and second halves of the record.

In this paper, we considered the ozone and temperature changes in the upper stratosphere using the re-analysis station data of NCEP/NCAR (National Centers for Environmental Prediction / National Center for Atmospheric Research) and JRA-25 (Japanese Re-Analysis 25 years). The main characteristics from 1992 to 2008 are as follows.

- (1) In the late spring (October and November) when the solar elevation angle in the Antarctic region is higher, the clear negative correlation between ozone and the monthly mean temperature variability at 100 hPa and 5 hPa is shown.
- (2) The monthly averages of the ozone (layer 8) and temperature (at 5 hPa atmospheric pressure level) for the month of November show high correlation (correlation coefficient is 0.9) between a decline of temperature, and the increase in ozone.
- (3) This suggests the relation of temperature dependence (about $2\%/1$ degree C) to the ozone change in the upper stratosphere (about 40 km in height).

The impact of the Montreal protocol is already seen in mid-latitude ozone, whereas the ozone change relevant to ODS in the upper stratosphere has not been observed due to the ODS still in transport to the Antarctic region.

GENERAL INTRODUCTION

The ozone hole was originally discovered by the Japanese researcher [Chubachi; 1984] in the 1980s, and depletion of ozone has accelerated rapidly after that. In 1974, Molina and Roland [1974] predicted depletion of ozone layer by the photochemical reaction with the man-made CFCs, which were transported to the stratosphere (height of about 40 km). In the upper stratosphere, ozone destruction occurs through the gas phase catalytic reaction with chlorine atom. On the other hand, ozone depletion in the lower stratosphere (height of about 15 km) takes place through the heterogeneous reaction on the surface of polar stratospheric cloud (PSCs: Polar Stratospheric Clouds) [Solomon *et al.*, 1986]. PSCs contain particles of water vapor (H₂O), nitric acid (HNO₃), and sulfuric acid (H₂SO₄), created under the specific climate condition (low temperature) in the Antarctic region. The ozone layer where the depletion was first discovered has recently shown a sign of recovery beginning in 1997 as a result of the Montreal Protocol initiated control of CFCs [Newchurch *et al.*, 2003]. Although the first stage of ozone recovery described as a slowdown of an ozone reduction has been already observed, the second stage of the statistically significant ozone increase has not yet been reached [WMO, 2007]. Even at the current stage of enhanced control of CFCs production, the Antarctic region shows a delay in ozone recovery as compared to the mid-low latitude trends. Therefore, good surveys of the continuous data in ground-based observations, especially observation of high quality are required. The major concern is that global warming will affect depletion of the ozone layer.

The Dobson ozone spectrophotometer was developed in the mid-1920s to measure stratospheric ozone, to assist in investigations of atmospheric circulation. The Dobson instrument and the method of operation are well documented [Dobson, 1931; Evans, 2008]. The Dobson's optical system is shown in Figure 1. The instrument measures the differential intensity of Solar light at selected wavelength pairs (traditionally called A, C, and D) in the ultraviolet spectral region. Table 1 shows ozone absorption and molecular scattering coefficients selected for Dobson measurements following the recommendation of the International Ozone Commission (IOC) in 1992. The molecular scattering coefficients β is proportional to a wavelength as $\lambda^{-4.27}$. The ozone absorption and molecular scattering coefficients for Total ozone and Umkehr retrieval algorithm are shown in Figure 2. Additional valuable information describing the accuracy of Dobson spectrophotometer observations is published by Dobson and Normand [1962]. The instruments remain the same optically, but advances in electronics and computer data acquisition and control have improved quality of data collection by automated instruments.

The Dobson ozone spectrophotometer is used in order to produce not only a measurement of total ozone amounts but an ozone vertical profile. The Umkehr technique has been used since the 1930s [Götz *et al.*, 1934] to estimate the vertical ozone profile from zenith sky measurements. The radiances are taken by the Dobson spectrophotometer [Dütsch, 1959] at a pair of wavelengths Ultra Violet wavelengths (C-pair band-passes are centered at 311.5 and 332.4 nm). Gradual changes in the radiance ratio with sun elevation including reversal of the slope at ~85 degrees solar zenith angles are called the “Umkehr curve”. A standard Umkehr observation consists of a series of C-pair wavelength measurements made under the

clear zenith sky conditions during a morning or an afternoon. The Umkehr curve measurements are related to the effective scattering height in the atmosphere that depends on the wavelengths at which observations are made (See Figure 3). In the standard Umkehr data for C wavelength pair processing the intensity NC value is recorded at specific solar zenith angles (60°, 65°, 70°, 74°, 75°, 77°, 80°, 83°, 84°, 85°, 86.5°, 88°, 89°, and 90°). The Umkehr technique has an advantage of not requiring the absolute radiation calibration in order to infer ozone profile information. The ground-based Umkehr method technique is very similar to the satellite backscatter ultraviolet (BUV) technique. Currently, measurements of the Umkehr observations are made routinely by automated Dobson spectrophotometers at a number of stations, primarily in the United States', Europe, and Japanese networks. Also, the JMA developed the international automation observation system, which is aimed at the long-term observation, maintaining stable and high quality data acquisition of Umkehr observation (See Annex B).

Table 1. Ozone absorption and molecular scattering coefficients for use with Dobson ozone Spectrophotometers beginning on 1 January 1992, where α and $\acute{\alpha}$ is ozone absorption cross-section recommended by the IOC letter in 1967.

Wavelengthpair	Slit	Wavelength $\lambda(\text{nm})$	Ozone Absorption $\alpha, \acute{\alpha} (\text{atm-cm})^{-1}$			Molecular Scattering coefficients $\beta, \beta' (\text{atm})^{-1}$			$(\beta - \beta') / (\alpha - \acute{\alpha})$ (atm-cm/atm)
			α	$\acute{\alpha}$	$\alpha - \acute{\alpha}$	β	β'	$\beta - \beta'$	
A	S2	305.5	(1.882)		1.806	0.489	0.114	0.063	
	S3	325.0	(0.120)			0.375			
B	S2	308.9	(1.287)		1.192	0.466	0.111	0.093	
	S3	329.1	(0.064)			0.355			
C	S2	311.5	(0.912)		0.833	0.450	0.109	0.131	
	S3	332.4	(0.047)			0.341			
D	S2	317.5	(0.391)		0.374	0.414	0.104	0.278	
	S3	339.9	(0.017)			0.310			

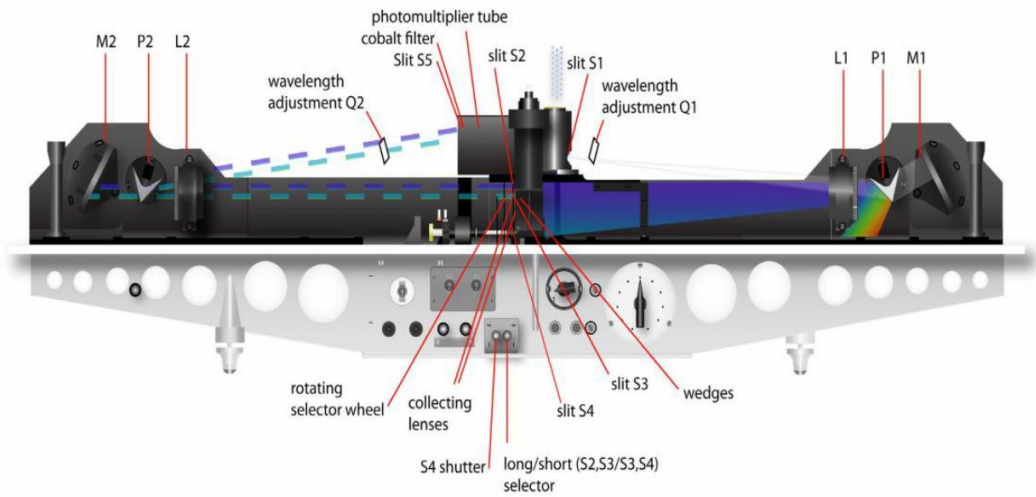


Figure 1. Optical system of the Dobson Spectrophotometer [Evans, 2008]

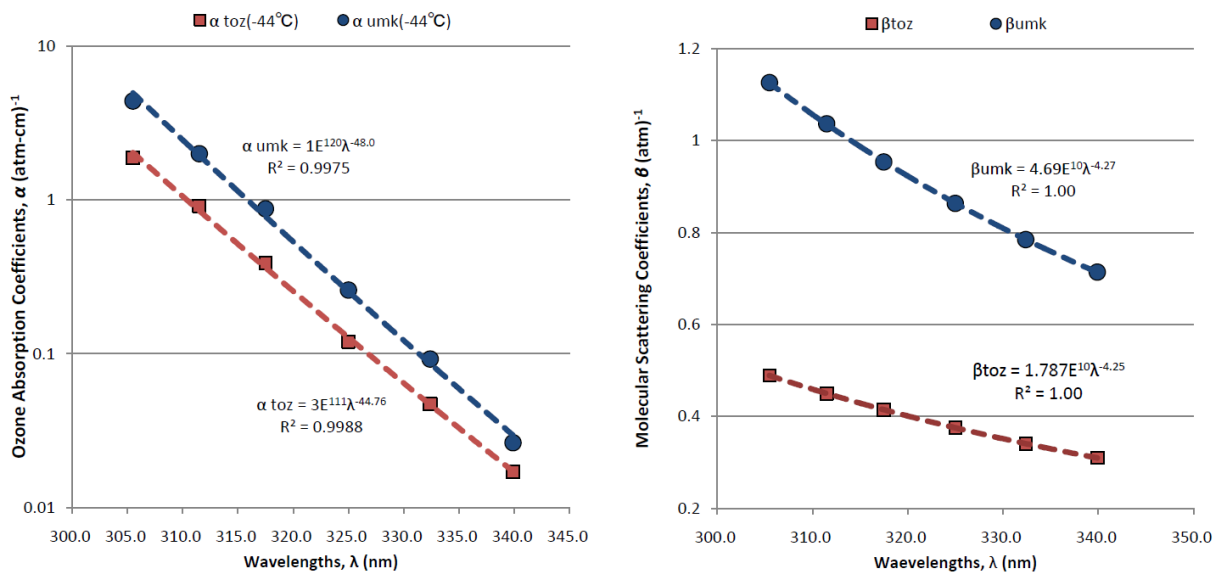


Figure 2. Ozone absorption and molecular scattering coefficients for the total ozone and Umkehr observation by Dobson Spectrophotometer.

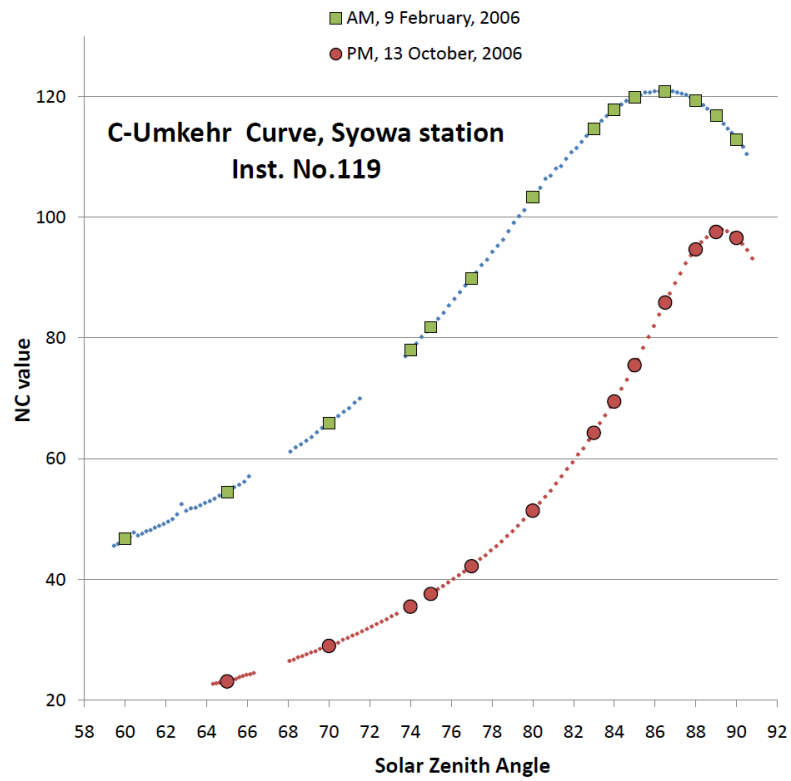


Figure 3. Umkehr C-curve taken from measurement

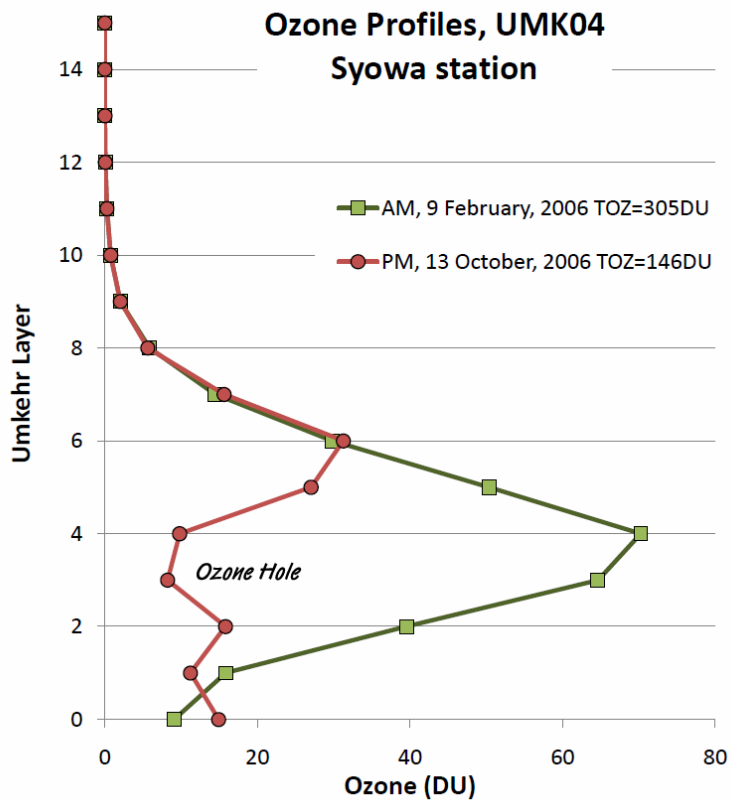


Figure 4. Umkehr retrieval ozone profile in 16-layers from Figure 3.

The long-term trend in ozone profiles is estimated for each station. Profiles are derived by the UMK04 algorithm [Petropavlovskikh *et al.*, 2005b]. The development of the Umkehr retrieval (UMK04) has played an important role for the monitoring of the SBUV derived ozone drifts and cross calibration of the BUV instrument [Fioletov *et al.*, 2006; Petropavlovskikh *et al.*, 2005a]. The forward model of the algorithm simulates Umkehr measurements based on the first-guess ozone profile, while inverse model uses a maximum likelihood estimation procedure and is based on Rodgers [2000], optimum estimation method, also used in the UMK92 retrieval [Mateer and DeLuisi, 1992]. The forward model is configured for 61 atmospheric layers with specified optical properties (including ozone absorption and molecular scattering) to simulate Umkehr curve. The ozone profile retrieval of the inverse method is also done at higher resolution, which is consistent with the vertical resolution of the forward model, and effectively eliminates interpolation errors embedded in the UMK92 code. The vertical profiles retrieved from two Umkehr measurements are shown as function of 16 Umkehr layers in Figure 4. The 61 and 16 layer systems are explained in Figure 5 and Table 2.

The Umkehr measurements, along with satellite and lidar measurements have robust information about ozone variability in the 40-km region, which is above the level at which the typical ozonesonde balloon bursts. Numerous studies have addressed accuracy of the ozone vertical distribution by Umkehr observation. The REVUE (REconstruction of Vertical ozone distributions from Umkehr Estimates) project of European Commission and World Meteorological Organization (WMO) was established to evaluate the role and results of Umkehr observations. In this project, continuity and consistency of Umkehr data of more than 40 years were analyzed [Godin *et al.*, 2000; Bojkov *et al.*, 2002]. Petropavlovskikh *et al.* [2001] showed that discontinuities found in Umkehr data worsen the quality of retrieved ozone profile and proposed the statistical way to correct shift error that also show SZA dependency. Using this method, Bojkov *et al.* [2002] reevaluated Umkehr data from many stations including Japanese stations. In Japanese Dobson network, a field instrument at each station had been frequently replaced by a different instrument in the network. Discontinuities or errors found in Umkehr data of Japanese network seem to be related to the difference in characterization of each instrument before and after replacement. Detailed information about each instrument replacement allows for appropriate correction of Japanese stations records [Bojkov *et al.*, 2002].

Comparisons of ozone profiles with different vertical resolution require application of a smoothing function to the highly resolved ozone profile to match the of lower resolution profile. The concept of Averaging Kernels (AK) was introduced by Rodgers [1976, 1990]. Figure 6 shows information content (or AK) for several individual and combined Umkehr layers. It is clear from the plot that vertical resolution for Umkehr retrieval is twice as large in the lower (0+1 and 2+3) or upper layers (8 and 9+10) as compared to the middle atmospheric layers (4, 5, 6, and 7).

Table 2. Layers used for Umkehr ozone profile retrievals [*Mateer and DeLuisi, 1992*]

Layers	Layer Base Approx. Height (km)	Layer Base Pressure (hPa)
1	0.0	1013.2
2	10.3	253.3
3	14.7	126.7
4	19.1	63.3
5	23.5	31.7
6	28	15.8
7	32.6	7.9
8	37.5	3.96
9	42.6	1.98
10	47.9	0.99

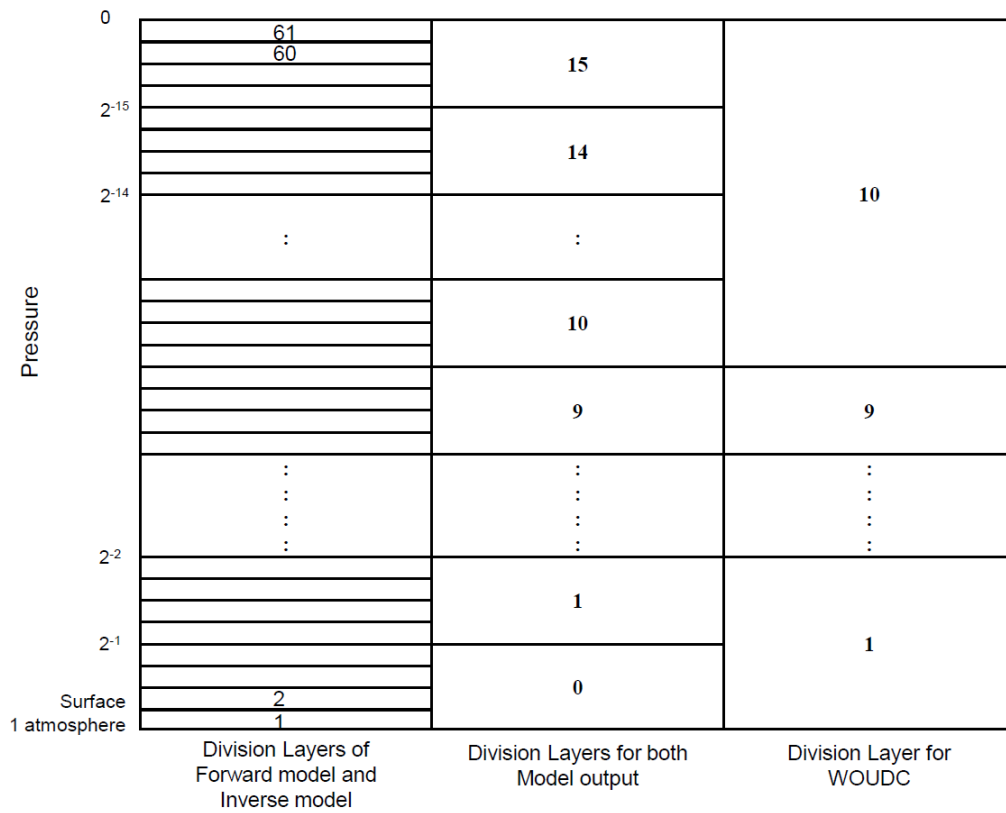


Figure 5. Division of the atmosphere by forward model and inverse model

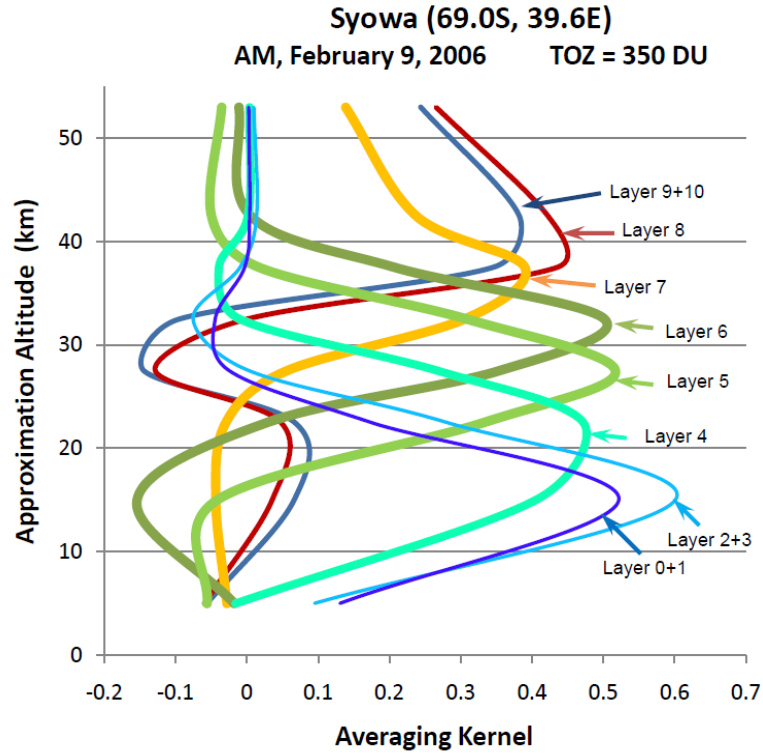


Figure 6. Each line shows algorithm's response (AK) to ozone change in layers

The ground-based ozone profile observations that use the Umkehr technique have been well established over the long history of ozone measurements, and have consistently provided data for the trend analysis [WMO, 2007]. However, Umkehr observations contain an issue that was uncovered by several comparison campaigns. Whenever multiple instruments take simultaneous Umkehr observations slightly different results are found that also depend on both the SZA and total ozone column. The contribution of the out-of-band stray light in the measurement becomes significant in case of the enhanced atmosphere extinction (high total ozone amount, low sun elevation) especially at the shorter wavelength in the pair. The short term variability in total ozone requires multiple observations for correction of SZA dependence in the stray light contribution. This is a time-consuming and expensive process. The measurement of stray light inside of a specific Dobson instrument can be used to develop the stray-light correction for that instrument, which could be applied to other, inter-compared instruments [Petropavlovskikh *et al.*, 2008, 2009]. The comparison of the measured N-value to a reference can be used to estimate the stray-light for an instrument, where the reference is simulated from auxiliary information from the ozone profile taken by co-incident and co-located ozonesonde and satellite observations. The improvement of several percent is shown in comparison of stray-light corrected Umkehr ozone profiles with ozonesonde in the troposphere and lower stratosphere [Petropavlovskikh *et al.*, 2009]. The stray-light model correction to the Umkehr observation network would require further research.

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Reevaluation of long-term Umkehr data and ozone profiles at Japanese stations

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[1] Umkehr observations have been routinely conducted at Japanese stations, Sapporo, Tsukuba, Kagoshima, and Naha, and the Antarctic station, Syowa, for more than 50 years. Umkehr data are a valuable source of information on long-term changes in the ozone vertical profile; however, the Umkehr record at Japanese stations has evident discontinuities. The majority of the discontinuities are related to the exchange of instruments for calibration (for total ozone measurements) and the replacement of instruments. These discontinuities may be related to the difference in instrument characteristics. In this article, reevaluation of the long-term Umkehr data in Japanese network is done by assessment of instrument-related changes in compared N values that exhibit solar zenith angle and total ozone dependence in addition to the step changes. The systematic errors are evaluated by simultaneous intercomparisons of each instrument with the reference instrument. Through this reevaluation, most discontinuities in a station's Umkehr time series are successfully corrected, and new sets of ozone vertical profiles are derived. The ozone profiles retrieved by two available Umkehr retrieval algorithms are compared with ozonesonde observations at every station and ozone lidar observations at Tsukuba. The results show that the revised Umkehr ozone profiles show improved consistency with both types of auxiliary ozone observations as compared to the old data sets, especially with regard to ozonesonde observations (difference of less than 5%). Trend analyses of the revised Umkehr ozone profile time series show a significant decrease in stratospheric ozone over Japan during the 1980s. It also varies between stations, with Naha showing the least significant trend among Japanese stations and Sapporo exhibiting as much as 6% of ozone decline per decade. In addition, a positive and statistically significant trend is detected in tropospheric ozone column at Naha (~5.5% per decade) and Tsukuba (~3.5% per decade) stations over the last 20 years, but no significant trend is observed over Sapporo.

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1. Introduction

[2] Among various data sources of vertical ozone distribution, Umkehr ozone observation records are important because they contain information over a wide range of altitudes and geographical areas and over a long time span [Reinsel, 2002; World Meteorological Organization, 2007; Fioletov *et al.*, 2006; Zanis *et al.*, 2006]. Ozonesonde observation provides much finer vertical resolution data, but the measurement is typically confined below 30 km or so [Fujimoto *et al.*, 2000; Deshler *et al.*, 2008]. The ozone sounding data

record began in the late 1960s, and at fewer stations [Komhyr, 1964]. Satellite observation provides homogeneous global data, but the period of most data records is confined to after the 1980s [Bhartia *et al.*, 1981, 1996, 2004]. Lidar and millimeter wave data have even shorter time records [Nakane *et al.*, 1993; Park *et al.*, 2006; Nakazato *et al.*, 2007]. Umkehr observations provide data at many stations where total ozone observation with Dobson instruments started in late 1950s [Dütsch, 1957, 1959]. Therefore, ozone profiles retrieved from Umkehr data cover the longest period of all observations and are vital to validation of other observation methods and numerical models that simulate and predict ozone changes. However, Umkehr ozone profile records are known to be impeded by the quality of individual Umkehr measurements, instrument stability, and continuity of measurements [Godin *et al.*, 2000; Bojkov *et al.*, 2002; Petropavlovskikh *et al.*, 2008].

[3] The REVUE (REconstruction of Vertical ozone distributions from Umkehr Estimates) project of European Commission and World Meteorological Organization (WMO)

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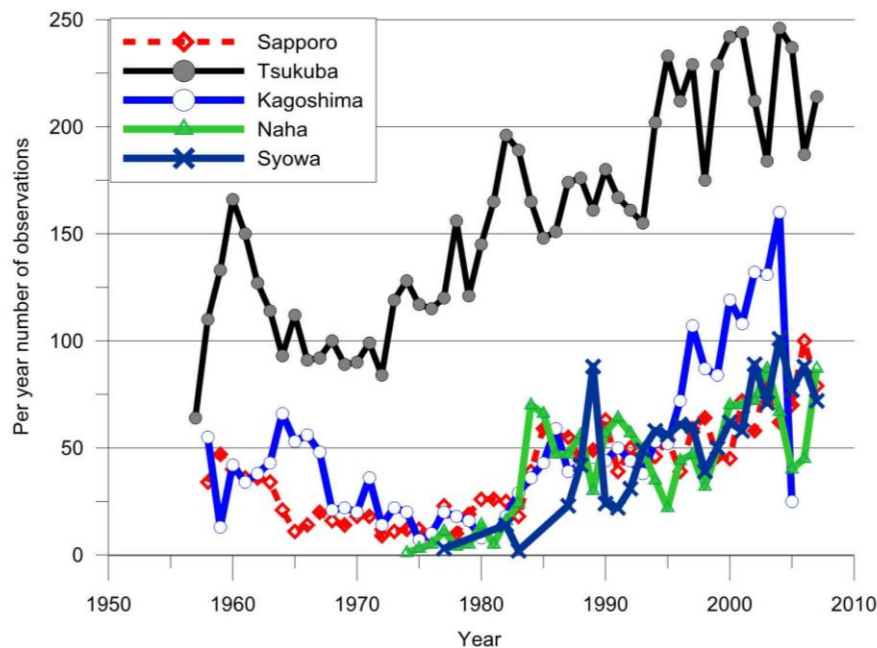


Figure 1. Per-year number of Umkehr observations in the Japanese observing network's history.

was established to evaluate the role and results of Umkehr observations. In this project, continuity and consistency of Umkehr data for more than 40 years were analyzed [Godin *et al.*, 2000]. Petropavlovskikh *et al.* [2001] pointed out that discontinuities found in Umkehr data worsen the quality of retrieved ozone profile and proposed a statistical way to correct shift error that depends on solar zenith angle (SZA). Using this method, Bojkov *et al.* [2002] reevaluated Umkehr data at many stations, including Japanese stations. In the Japanese Dobson network, a field instrument at each station had been frequently replaced by another instrument in the network. Discontinuities or errors found in Umkehr data of the Japanese network seem to relate to the difference in characterization of each instrument before and after replacement. With the full information about each instrument replacement, Bojkov *et al.* [2002] showed that appropriate correction can be made for Japanese stations.

[4] However, statistical correction can only be possible with a sufficient duration of measurements. Currently, calibration procedure for Umkehr observation has not been established. If a standard method is developed to assess the difference of field instruments to yield error in retrieved profile based on the instrument intercomparison data, the comparability and traceability of every instrument for Umkehr observation can be secured. Such a procedure was first studied by Miyagawa and Hirose [2004]. In this paper, a new correction method based on instrument intercomparison data and derived SZA and total ozone-dependent corrections is presented and applied to existing data sets to produce the revised data sets of Umkehr observation at Japanese stations.

[5] The new ozone profile retrieval algorithm, UMK04 [Petropavlovskikh *et al.*, 2005b], minimizes dependency of the retrieved ozone profile on total ozone changes, which is inherent in the former algorithm UMK92 [Mateer and

DeLuise, 1992]. Minimizing a priori information in the retrieval allows for an improved interpretation of trend analyses. Revised data sets of Japanese Umkehr observations are retrieved by both algorithms, UMK92 and UMK04, to examine the effects of new algorithm.

[6] In this study, after a brief introduction of the data set considered in section 2, a reevaluation of N values is performed by carefully considering various aspects of N values, which is described in section 3. Then in section 4, ozone profiles retrieved from reevaluated N values by using the UMK04 algorithm are examined and discussed by comparison with ozonesonde and lidar measurements. Conclusions are presented in section 5.

2. Data

[7] About 14,800 records of Umkehr observation data for Sapporo, Tsukuba (Tateno), Kagoshima, Naha and Syowa were collected since the Japan Meteorological Agency (JMA) began ozone observations in 1957 (see Table 1). The long-term records (through December 2007) of coincident Umkehr measurements at different Japanese stations are used for this study. Since 1994, the JMA has been acquiring Umkehr data of the standard Dobson C pair (311.5/332.4 nm) by a fully automated Dobson measuring system. Since July 2004, data of A and D pairs have been acquired in addition to C pairs. All three pairs of data are submitted to the World Ozone and UV Data Centre (WOUDC). The number of Umkehr records at each station was small except Tsukuba before 1984, but it has been steadily increasing since 1994 when the automated system was installed. Presently, about 230 records are acquired annually at Tsukuba station, which exceeds by 3 times the number of profiles acquired at other stations (see Figure 1). In this study, N values of C-pair

Table 1. Data Number of C-Umkehr Observation at Japanese Dobson Network^a

Station	Latitude	Longitude	Period	Number of Obs	Number of Months
Sapporo	43.1°N	141.3°E	1958–2007	1927	475
Tsukuba	36.1°N	140.1°E	1957–2007	7969	578
Kagoshima	31.6°N	130.6°E	1958–2005	2303	443
Naha	26.2°N	127.7°E	1974–2007	1398	323
Syowa	69.0°S	39.6°E	1977–2007	1240	167

^aNumber of obs and number of months indicate the number of individual observations and monthly mean values, respectively.

Umkehr measurements at 12 nominal SZAs (60°, 65°, 70°, 74°, 77°, 80°, 83°, 85°, 86.5°, 88°, 89°, and 90°) are used for analyses. With these N-value data, Umkehr algorithm allows derivation of 10-layer ozone amount. However, it has been pointed out that separation of information among some layers is actually difficult [Mateer, 1964, 1965; Mateer and DeLuisi, 1992]. In this study the Umkehr retrieved ozone profile layer system is chosen to separate vertical profile in layers 0 + 1, 2 + 3, 4, 5, 6, 7, and 8 and also 8 + 9 + 10 [Petropavlovskikh *et al.*, 2005b], where each Umkehr layer is ~5 km wide [Mateer and DeLuisi, 1992]. According to AK analysis this layer system is chosen to optimize ozone profile retrieval with uniformly distributed, a priori independent information on ozone variability in individual layers. The Umkehr method allows for separation of ozone column between layers 0 + 1 and 2 + 3 (~10-km-wide double Umkehr layers) though the quality of the lowermost layer 0 + 1 may be limited, whereas in satellite SBUV type measurements, the AKs analysis suggest combining layers 0, 1, 2, 3, and 4 [Bhartia *et al.*, 1996, 2004; Fioletov *et al.*, 2006].

[8] Data from the KC type ozonesondes are used for comparisons. The vertical resolution in sounding is typically on the order of 100 m and covers an altitude range below the burst level (variable, ~7 hPa pressure). The JMA network ozonesondes were routinely flown on a monthly basis before 1990 and weekly thereafter since 1968. Several models of KC ozonesonde have been used in analysis. (1) The model KC68 ozonesonde, introduced in 1968, was smaller and lighter than the first model KC65. (2) The model KC79, introduced in 1979, features up-to-date electronic techniques. (3) The latest model, KC96, introduced in 1997, applies new radiosonde unit, but the ozone sensor unit is the same as KC79.

[9] Data reduction algorithm for KC ozonesonde was recently revised to resolve some problems determined in the laboratory and field experiment campaigns such as JOSIE (Julich Ozone Sonde Intercomparison Experiment) and BESOS (Balloon Experiment on Standards for Ozone Sondes) [Fujimoto *et al.*, 2000; Deshler *et al.*, 2008].

[10] It was pointed out in the JOSIE campaign that KC ozonesondes tended to underestimate ozone in the lower atmosphere and overestimate it in the upper atmosphere [Smit and Kley, 1998]. Laboratory experiments revealed that this was due to solution features, such as response delay and temperature dependency of reaction current, and the new data reduction algorithm was introduced to correctly derive ozone concentration considering these effects. With new algorithm implementation, the Balloon Experiment on Standards for Ozone Sondes (BESOS) campaign shows that the accuracy of KC96 ozonesonde is improved to 5% in the middle and upper stratosphere, but it still underestimates the reference (photometer measurement) by over 10% in the lower atmo-

sphere [Deshler *et al.*, 2008]. Unresolved differences could be related to ozone loss through inlet tube and pump in early lifting stage.

[11] Ozone lidar measurements have been conducted at Tsukuba observational site by the National Institute for Environmental Studies (NIES) since 1988 under the Network for the Detection of Atmospheric Constituent Change (NDACC). The NIES system is composed of UV differential absorption lidar (DIAL) with a large telescope and high-power lasers. A new data processing and retrieval algorithm (version 2) has been recently introduced. Since the system installation in 1988, several replacements and improvements of parts have been made. The optical system of the lidar was altered in 1990 and 1995. Especially since 1995, accuracy in layers below 20 km altitude has been greatly improved [Park *et al.*, 2006]. For this study, we use the newest 553 profiles collected over the 1988–2007 period, as published in the NDACC site (<http://www.ndsc.ncep.gov/>), and unpublished data thereafter. National Centers for Environmental Prediction (NCEP) data sets and JMA's Global Analyses data sets are also used for altitude unit conversion. Ozone density by lidar is converted to ozone amount in each Umkehr layer by using the height and temperature information of NCEP data or the JMA's Global Analyses Data (24 layers from the surface to 0.4 hPa, above Umkehr layer 10).

3. Reevaluation of N Value

[12] The Japanese Dobson network has used nine instruments from Ealing-Beck Company and six from Shimadzu Company. Before 1994, ozone observation at each station was maintained by the periodical replacement of the field instruments by instruments which had been newly calibrated against the national standard 116 in Tsukuba. For example, at Sapporo, five Shimadzu and three Ealing instruments were deployed, and at Tsukuba, three Shimadzu and four Ealing instruments were used during 50 years (Figure 2).

[13] The instrument replacement seems to have often caused systematic errors and discontinuities in measurement readings due to the difference in optical characteristics (e.g., prism material, slit feature) of the succeeding instruments. Also, the calibration method used to assess and correct these effects of instrument-specific characteristics for Umkehr observation had not been yet established. In 1994, the Japanese Dobson network introduced a new system in which the calibration and maintenance are conducted at a field site using the traveling transfer standard, as well as the fully automated Dobson measurement system [Miyagawa, 1996]. Through this new system a single instrument is used at a site continuously. Thus the Umkehr data since this operational change have been found to be stable and of relatively good quality. However, if we analyze the long-term changes over

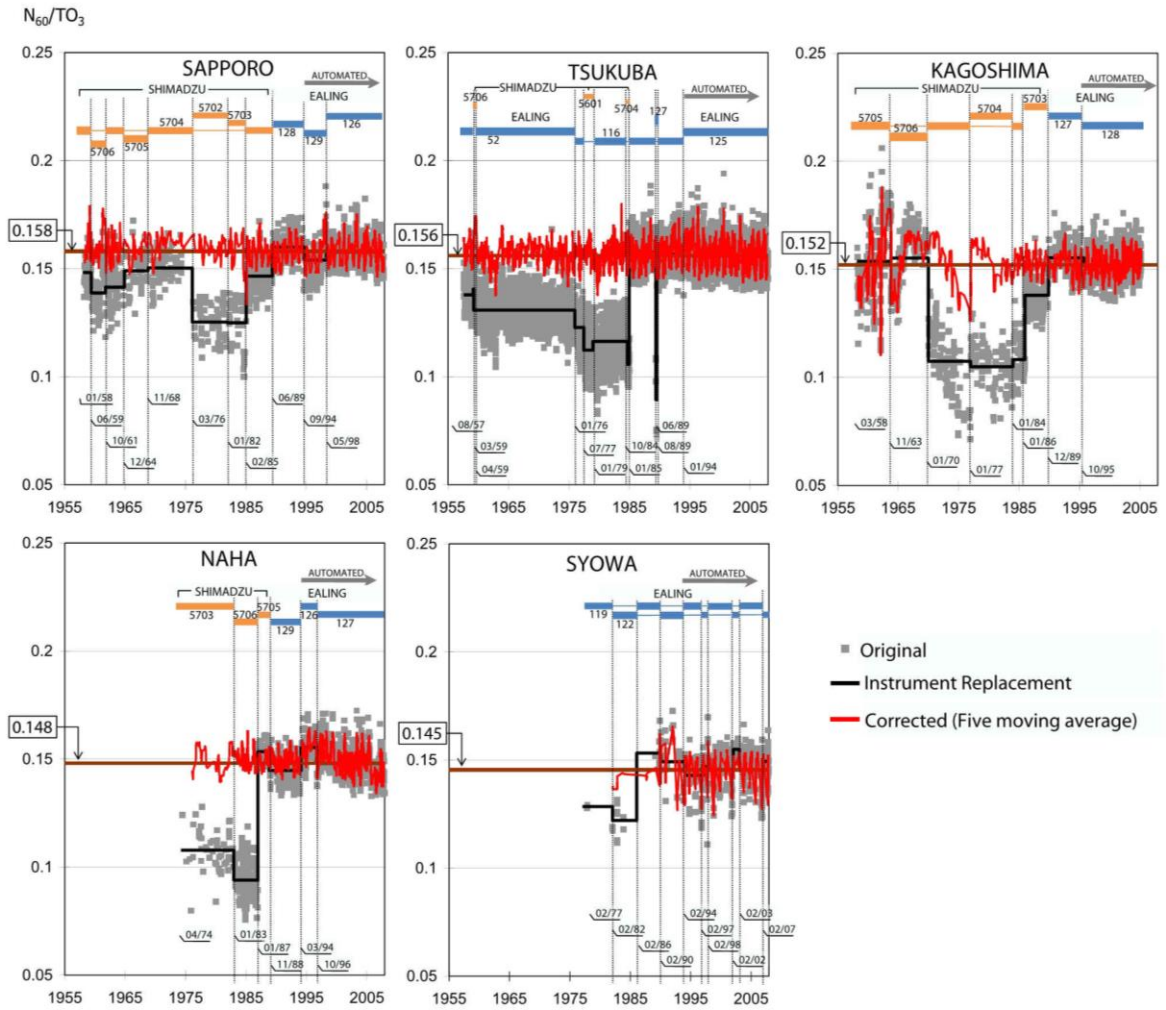


Figure 2. Ratio of the N value at 60° SZA to the total ozone (TO3) for each Umkehr measurement at Sapporo, Tsukuba, Kagoshima, Naha, and Syowa. Vertical dashed lines indicate events of instrument changes or calibrations, and deployed instruments between the events are shown by their instrument numbers with type (Shimadzu or Ealing) at the top of each plot. Month and year of the event is shown near the bottom of each panel. Horizontal black thick lines show average level in the period between events. Horizontal brown lines show average level between 1995 and 2007. The purple colored arrow indicates the period in which the measurement record was collected in the automated mode.

periods covered by several instruments, it is still necessary to assess systematic errors and discontinuities related to the instrument replacement. Though a statistical way to correct for the N-value errors and discontinuities related to the change on instruments in the measurement history of the Japanese stations was proposed by *Bojkov et al.* [2002], the method in this study relies more strongly on corrections based on the individual instrument characteristics. A comparison with the *Bojkov et al.* [2002] method would be the subject of a study after the development of a more complete general method for evaluating the individual instrument optical characteristics with respect to measurements of the Umkehr effect [*Evans et al.*, 2009; *Petropavlovskikh et al.*, 2008].

[14] In the Umkehr measurements a ratio in zenith-sky UV intensities at two wavelengths are utilized for ozone profile retrieval. Intensity ratio, N value, is written as

$$N(x, z) = 100 \log_{10} \left\{ \frac{[I(x, z, L2)/F_0(L2)]/[I(x, z, L1)/F_0(L1)]}{\dots} \right\} + k_0, \quad (1)$$

where $I(x, z, L)$ is intensity at wavelength L , x is the parameter of ozone profile, z is SZA, $F_0(L)$ is the extraterrestrial solar flux constant at wavelength L , and k_0 is the instrumental constant [*Mateer and DeLuisi*, 1992; *Evans*, 2006]. For C-pair measurement L1 and L2 are centered at 311.5 nm and 332.4 nm, respectively. For reevaluation of long-term

Umkehr data, the following N-value errors have to be assessed, i.e., (1) an error that appears as a step change in time series (shift error), (2) an error that depends on SZA (SZA error), and (3) an error that depends on total ozone in addition to SZA (total ozone error).

3.1. Shift Correction of N Value

[15] In the long-term N-value data, apparent shifts (discontinuous steps) are found corresponding to the date of instrument replacement. To eliminate such discontinuous steps in N value, a shift correction by statistical intervention was proposed [Bojkov *et al.*, 2002]. As N values are roughly proportional to total ozone, step-like discontinuity can be easily detected in time series as abrupt changes in the ratio of measured N value and total ozone, $NPTO_3$ (= N value/total ozone). Time series of $NPTO_3$, at SZA 60° , are shown in Figure 2 by gray dots for Sapporo, Tsukuba, Kagoshima, Naha, and Syowa for entire period of observation till 2007. Discontinuous steps, especially large prior to 1980s, can be easily identified at every station. Vertical lines in Figure 2 show the timing of instrument replacements. The operated instrument with type (Shimadzu or Ealing) is shown at the top of each plot. It is found that steps generally correspond to the instrument replacements. Horizontal black thick lines in Figure 2 show average level in the period between replacements. Horizontal brown lines show the average level between 1995 and 2007 (for the period of stable data acquisition by automated system).

[16] While a shift in the reevaluation by Bojkov *et al.* [2002] is assessed at each SZA, a shift in this study is assessed at only SZA 60° , which is usually used as the reference SZA in Umkehr retrieval procedure. In the algorithm to retrieve ozone profile, N values are analyzed after subtracting the N value at SZA 60° (or the highest available SZA) from measurements at other SZAs to remove uncertainties associated with the unknown instrumental constant (k_0). Thus, if a shift in N value is uniform in SZA, the effects of a shift will be canceled out. Thus a shift correction here has no effect on improvement of ozone profile, but serves only to homogenize time series data to remove any apparent discontinuities. Contrary to the previous studies, the SZA-dependent correction that has an effect on the retrieved profile is applied separately, after the shift correction, and at the later stage.

[17] First, a shift correction is assessed for each time period associated with instrument replacements. The shift $\Delta N_{\text{sft}}(t)$ of a period t is given by following formula with reference period (1995 ~ 2007).

$$\Delta N_{\text{sft}}(t) = [NPTO_{\text{ref}} - NPTO(t)]TO_{3\text{ref}} \quad (2)$$

$$N'(x, z, t) = N(x, z, t) + \Delta N_{\text{sft}}(t), \quad (3)$$

where $NPTO(t)$ is the average of N_{60}/TO_3 in the correction period t , $NPTO_{\text{ref}}$ is its value for the reference period, and $TO_{3\text{ref}}$ is the average of total ozone for the reference period. Shift correction $\Delta N_{\text{sft}}(t)$ is uniformly added to measured N values for all SZA through the period t to create revised N values, $N'(x, z, t)$.

[18] After shift correction, the steps in $NPTO_3$ are generally removed at all five stations, except for the period between the

beginning of the time series and 1976 at Kagoshima where small discontinuous steps still remain after correction. Effects of SO_2 emission of volcano Sakurajima nearby may be a possible cause [Komhyr and Evans, 1980; De Muer and De Backer, 1992]. The values of $NPTO_{\text{ref}}$ and $TO_{3\text{ref}}$ at the five stations are summarized in Figure 2.

3.2. SZA Correction of N Value

[19] N values in the $60^\circ \sim 90^\circ$ range of SZA are known to have systematic SZA depending biases that are specific for each instrument. These features are apparent in the results of some intercomparison campaigns, and are considered to be related to differences in the material and quality of individual instrument prisms and to effects of scattered light in the instrument [Petropavlovskikh *et al.*, 2004]. Bojkov *et al.* [2002] assessed SZA-dependent biases as a component of a shift error, but the systematic SZA-dependent biases can be evaluated by intercomparison between instruments. N-value differences between Japanese Dobson network instruments are derived from intercomparison data (see Figure 3). For the evaluation, we used 116 as the reference instrument since the world reference instrument for Umkehr measurements has yet to be established. Results of three Dobson intercomparison campaigns (Mauna Loa, 2001; Boulder, 2004 and 2007) of the world primary standard Dobson (083) and the Asian regional standard Dobson 116 show good consistency between the two instruments. The difference of N values in the range of $60^\circ \sim 90^\circ$ SZAs is found to be less than 0.4 (Figure 3a), which suggests its adequacy for a choice of the reference.

[20] In order to reduce measurement uncertainties, SZA-dependent biases are evaluated by means of averaging of the multiple intercomparison results. However, instruments used at the beginning of the record do not have sufficient data for statistically meaningful intercomparisons. Especially, four out of six Japanese Dobsons (Shimadzu Co.) have no such data as these have been already destroyed. Therefore, there is no easy way to obtain quantitative intercomparison data.

[21] Derived biases of 10 field instruments (8 Ealing and 2 Shimadzu) used in the Japanese network as well as the world primary standard 083 are shown in Figure 3a with reference to Dobson 116 coincident measurements. Dobson 52 is currently installed in Manila, but is shown here as operated in Tsukuba during 19 years from 1957. The result shows that Dobsons 52, 83, 119, and 122 have biases of 1 N, but that Dobsons 125 ~ 129 have systematically large biases as large as 4 N values at SZA larger than 80° . The relative biases between two Shimadzu Dobsons and Dobson 116 are about 2 N values, where the results are based on statistically significant data sets. Unfortunately, there is no information to derive meaningful biases for the other four destroyed Shimadzu instruments, but it is expected that their biases are in the same range as the one that is discussed above.

[22] In Asia, Dobson intercomparisons to Asian standard 116 were conducted several times. Figure 3b shows biases of Asian network instruments derived by simultaneous Umkehr measurements. Biases of less than 1 N value were found when the instruments 52, 90, 100, 109, and 112 were compared against 116 during the campaigns of 1996, 2003, and 2006. On the contrary, intercomparison of 124 (Seoul, Korea) in 2004 and 2005 show large biases that are similar to 125 ~ 129 results. Therefore, Dobson instruments with numbers

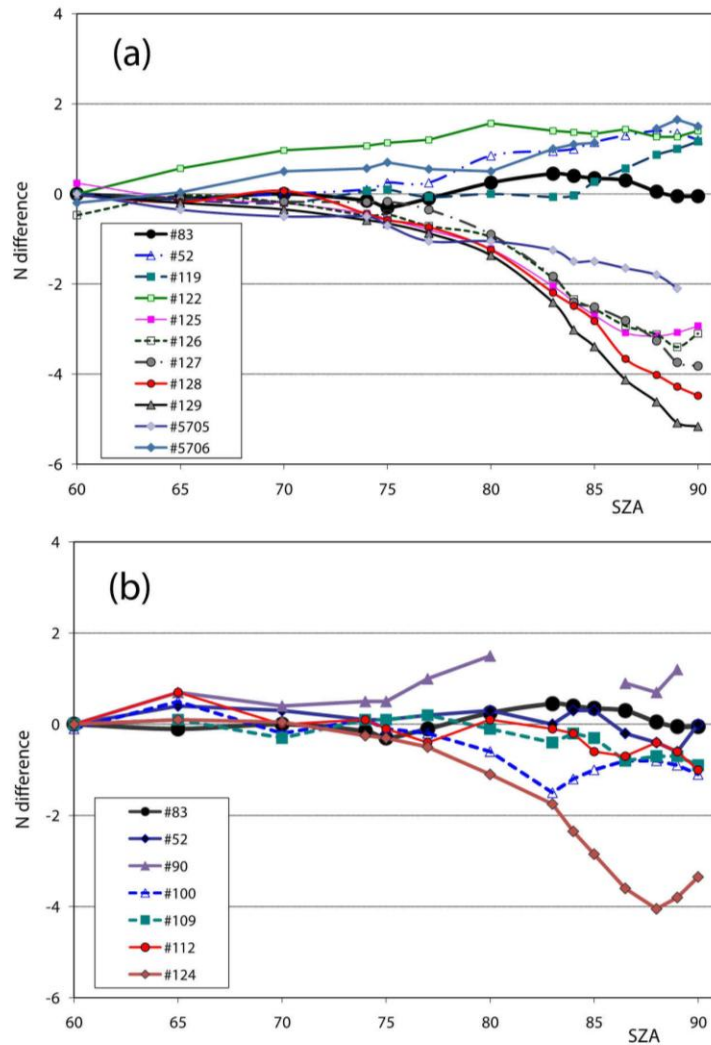


Figure 3. SZA-dependent N-value biases based on the intercomparison of Umkehr measurements. (a) Biases of the world primary standard Dobson 83 (thick line) and of field instruments in the Japanese network in relation to Asian regional standard Dobson 116. (b) Biases of instruments of the Asian network referred to 116.

124 and larger seem to have different characteristics from the Dobsons with numbers smaller than 124. This may be due to the change in optical parts of Dobson as pointed out by *Evans et al.* [2009].

[23] SZA-dependent bias is evaluated as the difference of N-value readings between the analyzed instrument and the reference instrument, such that SZA correction $\Delta N_z(z, \text{ins})$ is described by (4), and SZA corrected N-value N'' are defined by (5).

$$\Delta N_z(z, \text{ins}) = \frac{\sum \{ [N(z, \text{ref_ins}) - N(60, \text{ref_ins})] - [N(z, \text{ins}) - N(60, \text{ins})] \}}{n} \quad (4)$$

$$N''(x, z, t, \text{ins}) = N'(x, z, t) + \Delta N_z(z, \text{ins}). \quad (5)$$

In (4), the parameter “ins” means the analyzed instrument, “ref_ins” means the reference instrument, and n is the number of intercomparisons. Summation is done n times. In this study the Asian regional standard Dobson 116 is selected as the reference instrument. If an instrument has no direct intercomparison data with the reference instrument, SZA correction is determined by indirect intercomparison using the traveling standard instrument 129.

[24] Contribution of SZA-dependent bias to error in ozone profile is estimated at 20% in layers 7 and 8. For four Shimadzu instruments that have no intercomparison data, no SZA correction was applied. If SZA errors of these Shimadzu instruments are on the same order of other Shimadzu instruments, about 5% uncertainty is estimated to be contained in the retrieved profile for the period when these instruments were used.

3.3. Total Ozone Correction of N Value

[25] After SZA correction, N values have still discernible uncertainty errors. Some of these errors have total ozone (TO) dependent nature, which is suggested by results of the inter-comparison campaign by *Petropavlovskikh et al.* [2004] and is quantitatively discussed by *Miyagawa and Hirose* [2004].

[26] In the period between 1998 and 2004, a significant number of Umkehr intercomparisons were executed between the Asian regional standard 116 and the field instruments 125 through 129 at Tsukuba. The remaining error of 125 after shift correction and SZA correction in reference to 116 has total ozone dependency of about 1.7 N values at SZA 84° per 100 DU total ozone (Figure 4a). TO dependency is especially large in N values at SZA between 77° and 90° where zenith scattered light is weak (Figure 4b). Similar TO dependency is found for Dobson instruments 126 through 129. To demonstrate the instrumental characteristics of derived corrections, results for 129 are also plotted in Figures 4a and 4b in addition to 125. After total ozone correction, the uncertainty error for N values has been reduced by about 30%. The range of error bars before and after total ozone correction is shown in Figure 4c. Total ozone correction $\Delta N_{to}(to3, z, ins)$ can be expressed by

$$\Delta N_{to}(to3, z, ins) = \alpha(z, ins) + TO3 \beta(z, ins), \quad (6)$$

where coefficients α and β are the intercept and slope, respectively, when the difference between the N values of the analyzed instrument (ins) and the reference instrument (ref_ins) is expressed as linear function of total ozone TO3. Adding the term $\Delta N_{to}(TO3, z, ins)$ to $N''(x, z, t, ins)$ derived in (5), we can obtain corrected N value $N'''(x, z, t, ins, TO3)$ as follows:

$$N'''(x, z, t, ins, TO3) = N''(x, z, t, ins) + \Delta N_{to}(TO3, z, ins). \quad (7)$$

To make a total ozone correction a significant number of intercomparisons with the reference is needed to derive reliable regression coefficients. Therefore, total ozone corrections are applied only to measurements by Dobsons 125 through 129 individually, and are not applied to the other instruments in this study. Contribution of total ozone-dependent bias to error in ozone profile is estimated to be at most 5%.

4. Validation of Retrieved Ozone Profile

[27] Ozone profiles were retrieved from N values after the corrections described above were implemented into the time series at each station. The newly reprocessed Umkehr ozone profiles are validated by comparisons with ozone profiles

from independent methods, i.e., ozonesonde and ozone lidar. Umkehr ozone profiles are retrieved by both the traditional algorithm (UMK92) and the new algorithm (UMK04) to assess the algorithm-related differences. In UMK92, data are processed with the reference chosen at 60° SZA (or the smallest SZA available below 70°), while in UMK04 data are processed with normalization fixed at 70° SZA.

[28] Figures 4d, 4e, and 4f demonstrate how the SZA and/or TO corrections change the retrieved ozone for Tsukuba station. Figure 4d shows the percent changes in Umkehr layers 8, 4, and 1 ozone due to SZA correction of N values in the Tsukuba record as a function of time. Figure 4e demonstrates layer 8 ozone changes due to TO correction. Finally, Figure 4f demonstrates the combined effect of SZA and TO corrections on layer 8 ozone at Tsukuba station. In this case a large change in ozone amount over 20% (from 1975 to 1994) is found mainly by SZA correction. Especially the field instrument replacement from 116 to 125 in 1994 brings about a large change in retrieved ozone by SZA correction. On the basis of the characteristic difference in SZA dependency (Figure 3), instruments with numbers 124 or larger and numbers smaller than 124 are considered to make large differences in retrieved ozone by SZA correction.

4.1. Effects of N Value Reevaluation on Derived Ozone Profiles

[29] The ozone profiles were retrieved with the UMK04 first with the original (ORG) and then revised (REV) set of N values at Sapporo, Tsukuba, and Naha stations. Figure 5 shows the percent differences of annually averaged layer ozone amount between the two data sets. Substantial differences are noticed. It is found that overestimated ozone on the order of 10–20% in layers 7, 8, and 9 after 1989 is corrected in the revised data. Conversely, underestimated ozone is found in layer 4 in the original data after 1989. These periods of overestimated and underestimated ozone are coincident with the use of instruments 125 ~ 129. Other corrections are on the order of about 5%. The zero period of the difference applied neither SZA correction nor TO correction. For example, correction in the period using 116 from 1976 to 1993 (except June–July 1989, using 127) shows the difference as zero in Tsukuba.

4.2. Effects of the Retrieval Algorithm on Derived Ozone Profiles

[30] The effect of the retrieval algorithm on the revised ozone profile time series was assessed through comparisons of UMK04 and UMK92 results. Figure 6 shows time series of retrieved ozone amount in layers 1, 4, and 8 at three Japanese stations. Ozone amount is retrieved by four procedures, i.e., combination of original (ORG) and revised (REV) N-value data sets, and UMK92 and UMK04 algorithms. First it is noticed that results by UMK92 and UMK04 are substantially

Figure 4. Mean of the difference of N values measured in simultaneous comparison with Dobson 116 and 125 (dark squares), and 116 and 129 (red dots). (a) The 60°-normalized N values at 84° SZA are plotted as a function of total ozone column. (b) Total ozone regression coefficients for N values (as presented in Figure 4a) are plotted as function of SZA. (c) Difference between 116 and 125 are plotted as a function of SZA for all TO values. Vertical bar shows the standard deviation after correction of total ozone, and dashed line shows uncorrected standard deviation. (d) Difference of retrieved ozone from original N values and SZA corrected N values at Tsukuba in layers 1, 4, and 8. (e) Difference of retrieved ozone from TO corrected N values at Tsukuba in layer 8. (f) Cumulative difference of retrieved ozone from original N values and SZA and TO corrected N values at Tsukuba in layer 8.

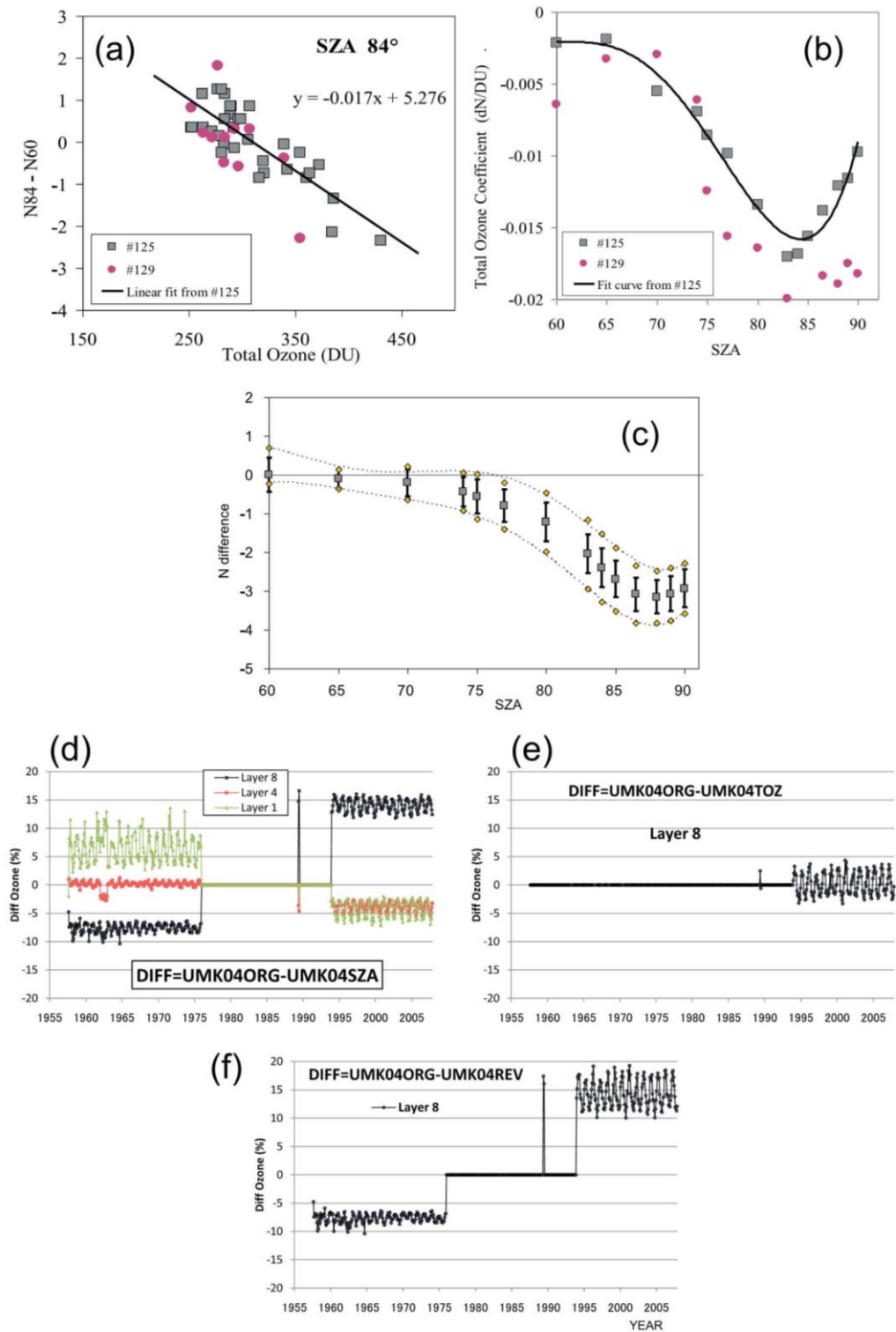


Figure 4

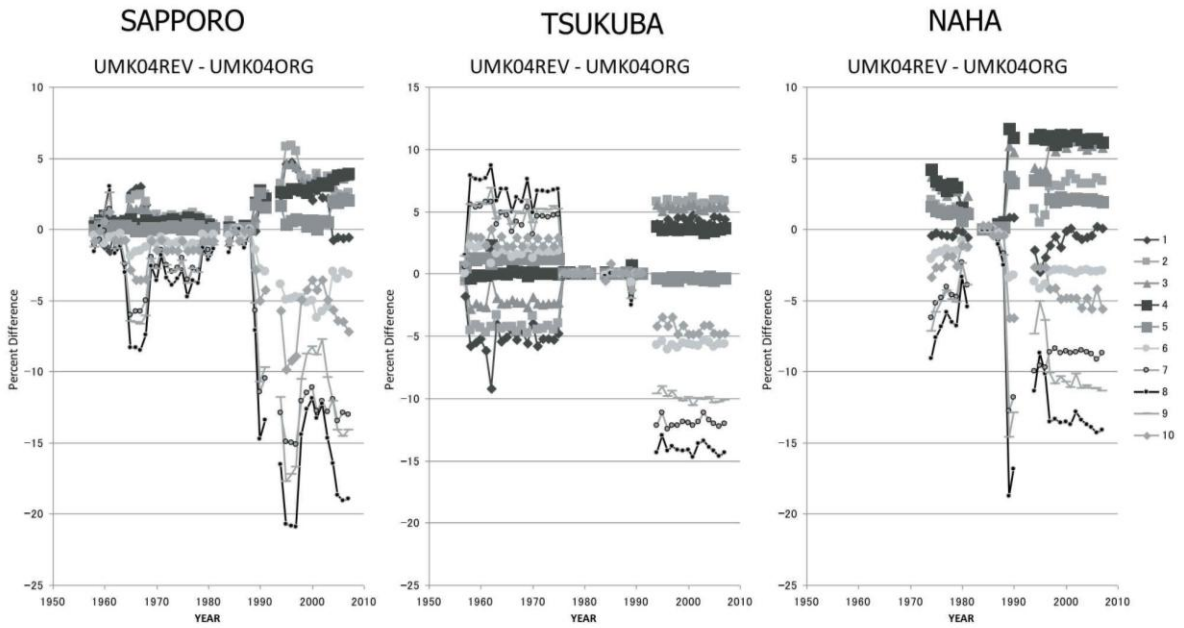


Figure 5. Difference between retrieved ozone when using the original (UMK04ORG) and revised (UMK04REV) algorithms for three stations is shown as a time series of offsets for individual Umkehr layers.

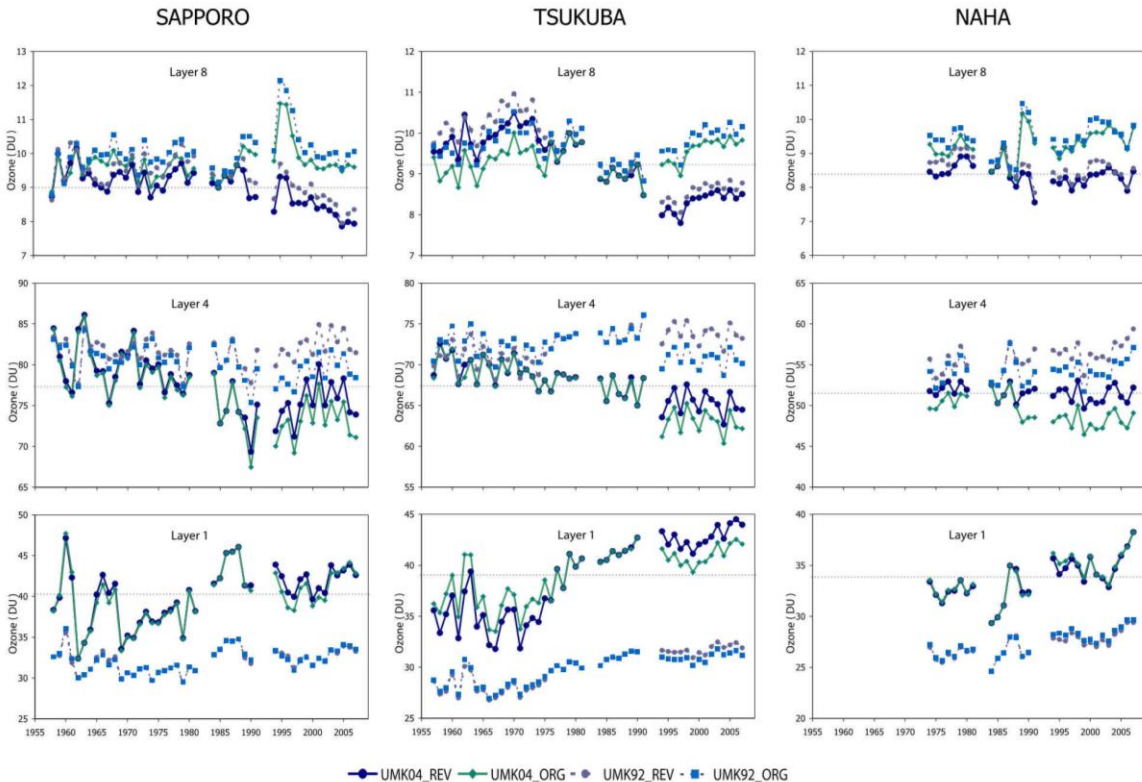


Figure 6. Retrieved ozone amounts in three layers by four types of procedures, i.e., combination of UMK92 and UMK04 algorithm from original (ORG) and revised (REV) N values, at three stations are shown. Dashed line shows the average ozone between 1960 and 2007 for UMK04_REV.

different, especially in layer 4. Though a clear decreasing trend between 1980 and the mid-1990s can be seen in UMK04 results, such a tendency cannot be seen in UMK92 results. This is due to the total ozone dependency inherent in the UMK92 algorithm. The difference between both algorithms is small in layer 8 and is reversed in layer 1. In layer 1 the amount by UMK04 is larger than that by UMK92 through the analysis period by about 15%.

[31] As for the effects of Umkehr data revision, the downward correction in layer 8 in REV data after 1989 is remarkable. Accordingly, somewhat unexplained deviation in layer 8 ozone amount during 1990s is reduced. This seems consistent with the result of *Reinsel* [2002]. On the contrary, fairly low ozone levels in layer 4 at Tsukuba station after the mid-1990s are increased in both retrieval versions.

4.3. Comparison With Ozonesonde and With Ozone Lidar

[32] Retrieved ozone profiles by UMK04 are compared with ozonesonde profiles and ozone lidar profiles. The scheme of Umkehr averaging kernel is used to make the layer averages easy to compare. As the vertical resolution of Umkehr retrieved ozone profiles is limited [*Petropavlovskikh et al.*, 2005b], all ozonesonde profiles are converted to Umkehr layers 0 + 1, 2 + 3, 4, 5, and 6. Layer 7 is also used for comparison with lidar, which provides data up to higher altitudes. The coincidence limit for compared data sets is set to 1 day maximum. Ozone density by lidar is converted to ozone amount in each Umkehr layer by using the height and temperature information of NCEP data or the JMA's Global Analyses Data (24 layers from the surface to 0.4 hPa, above Umkehr layer 10). In addition, the coincident Averaging Kernel (AK) that is derived in the Umkehr algorithm is applied for smoothing of the vertically resolved ozonesonde and lidar profiles. To appropriately apply algorithm smoothing in each layer, UMK04 AKs and Umkehr a priori ozone profiles are combined in 16 single Umkehr layers, where atmospheric pressure at each layer's top level is half of the pressure at its bottom. In order to simulate the Umkehr-smoothed ozonesonde or lidar profile information, $X_{sm}(j)$, the following procedure is followed:

$$X_{sm}(j) = \sum_k \{AK(j, k) \times [X_i(k) - AP(k)]\} + AP(j),$$

where j is the Umkehr layer number, the $X_i(k)$ is 16-layer ozone sounding or lidar profile in layer k , $AP(k)$ is Umkehr a priori in layer k , and \sum_k is the integral of the smoothed differences in all 16 layers. If the ozone sounding or lidar data are not available in the k th layer, the $[X_i(k) - AP(k)]$ difference is set to zero.

4.4. Comparison in Time Series

[33] Figure 7a shows comparisons of monthly averages of ozonesonde and Umkehr profiles taken on the same day at Tsukuba and Syowa stations. Results are shown for ozone time series in layers 0 + 1, 2 + 3, 4, 5, and 6. Both ozone amounts by Umkehr and ozone sounding, and the respective differences (%) are shown, where differences are defined as

$$\text{Percent Diff}(ij) = (O_{ij} - U_{ij}) / \text{Average}(MO_j + MU_j) \times 100.$$

(8) ¹Auxiliary materials are available in the HTML. doi:10.1029/2008JD010658.

Here i and j mean year and month, respectively, and O_{ij} and U_{ij} are monthly averaged ozone amount of month j of year i by ozonesonde and by Umkehr measurement, respectively. MO_j and MU_j represent the long-term average ozone for ozonesonde and Umkehr in month j . Both data sets, especially with respect to seasonal variations, show similar ozone variability in all layers. However, in the troposphere (layer 0 + 1), ozone amount derived by ozonesonde is consistently lower than Umkehr-derived ozone.

[34] As was discussed before, the accuracy of KC96 ozonesonde is improved by a new algorithm within 5% in the middle and upper stratosphere, but it still underestimates by more than 10% in the lower atmosphere [*Deshler et al.*, 2008]. Thus, this lower amount by ozonesonde in layer 0 + 1 is consistent with the KC ozonesonde's characteristics, though the accuracy of Umkehr layer 0 + 1 may also be somewhat limited. In layers 2 + 3 and above, both results are generally consistent and residual scattering is relatively small.

[35] Elevated difference between the two data sets and larger residuals can be seen around 1992 at higher altitudes (layers 5 and 6). This is related to the interference of the volcanic aerosols injected into the stratosphere by the eruption of Mount Pinatubo (June 1991) with Umkehr measurements, thus causing the errors in the Umkehr retrieved ozone profiles [*Petropavlovskikh et al.*, 2005a]. For ozonesonde and Umkehr comparison at other stations (Sapporo, Kagoshima and Naha), see the auxiliary material.¹

[36] Figure 7b shows monthly averages of coincident ozone amount measured by lidar and Umkehr. Comparison is done in Umkehr layers 3, 4, 5, 6, and 7. The seasonal variations are similar, but lidar results are generally higher than Umkehr results. Elevated difference between two measurements can also be seen around 1992 in layers 5, 6, and 7. As mentioned above, stratospheric aerosols are known to cause changes in N-value measurement and produce errors in the Umkehr ozone retrievals.

[37] Figure 8 shows the averaged seasonal variations of ozonesonde, Umkehr ozone and their difference (Figure 8, top), and lidar, Umkehr ozone and their difference (Figure 8, bottom) for each layer. Generally, seasonal variations are similar, and in higher layers, differences are small and do not show any seasonal dependency. However, in lower layers the differences between ozonesonde and Umkehr results are on the order of 20% for middle latitude sites and even larger over the Antarctica site. Generally ozonesonde results are lower than Umkehr results in layer 0 + 1, and its difference is the largest in summer months, except at Naha. At Syowa, as the ozone amount is low especially during the Austral spring with associated ozone depletion events, and the number of collected profiles is small, month-to-month ozone variability is relatively large. For lidar and Umkehr comparison, the month-to-month differences are similar across the year, but lidar ozone is somewhat larger than Umkehr ozone in layers 4 and above. See Table 2 for difference, standard deviation, and number of used data.

[38] Figure 9 shows differences found between the three methods as a function of layer and time. In lower layers (below layer 3) after 1990, ozonesonde data are generally

(a) SONDE vs UMKEHR

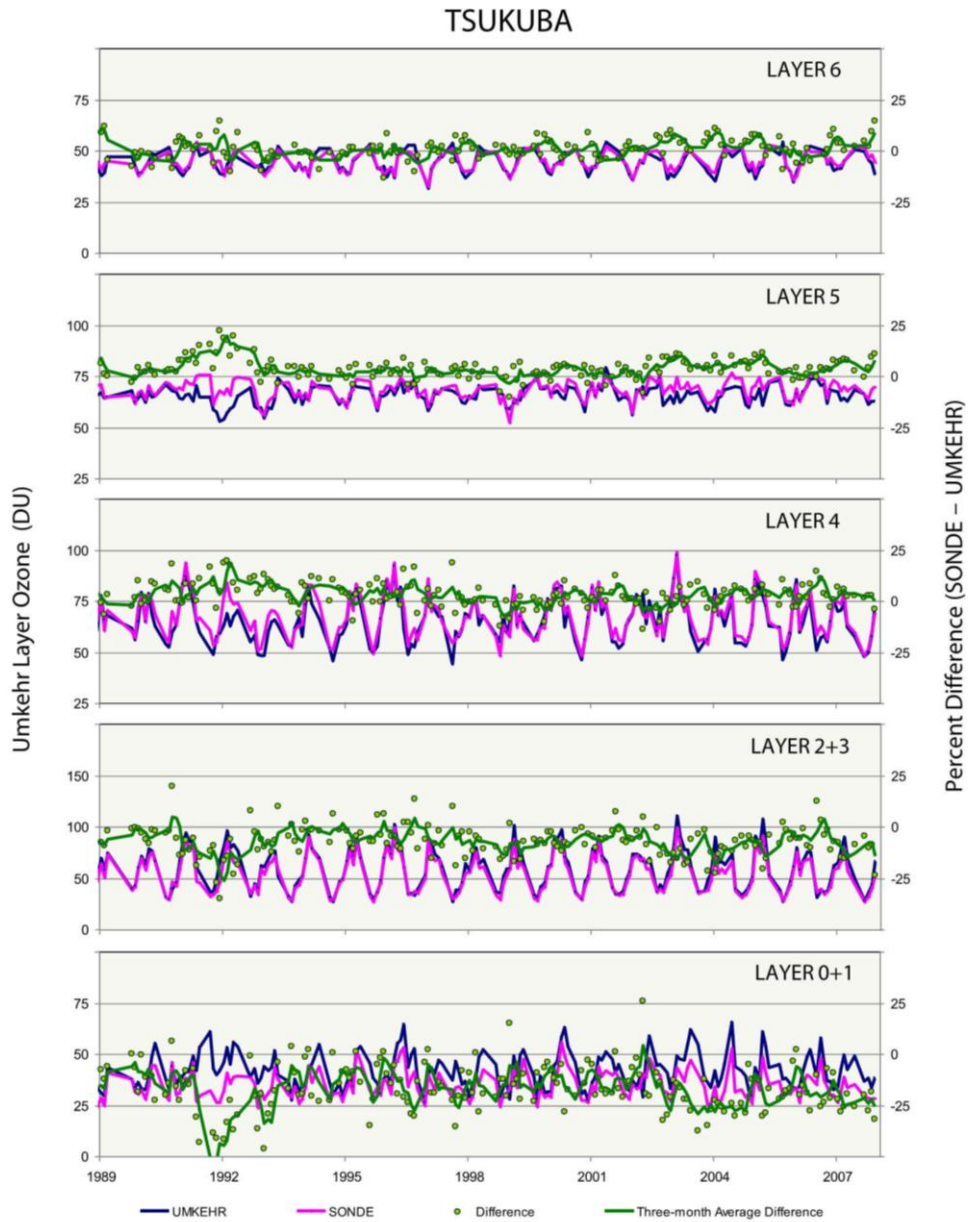


Figure 7. Time series of comparison of Umkehr with ozonesonde and with lidar. (a) Monthly average of daily comparison of ozone amount by ozonesonde and Umkehr measurement and their difference at two stations (Tsukuba and Syowa). (b) Monthly average of daily comparison of ozone amount by lidar and Umkehr measurement and their difference at Tsukuba.

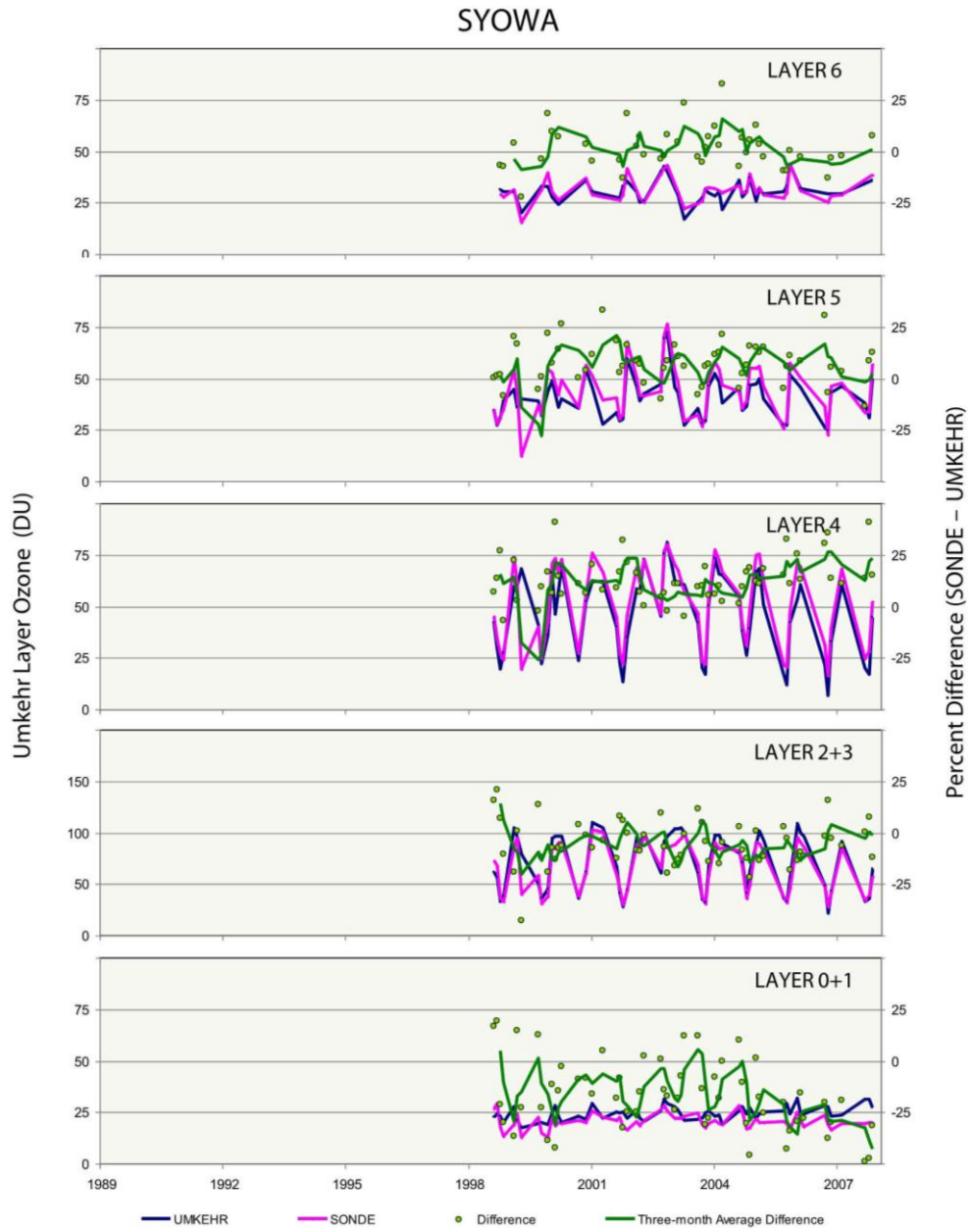


Figure 7. (continued)

(b) LIDAR vs UMKEHR

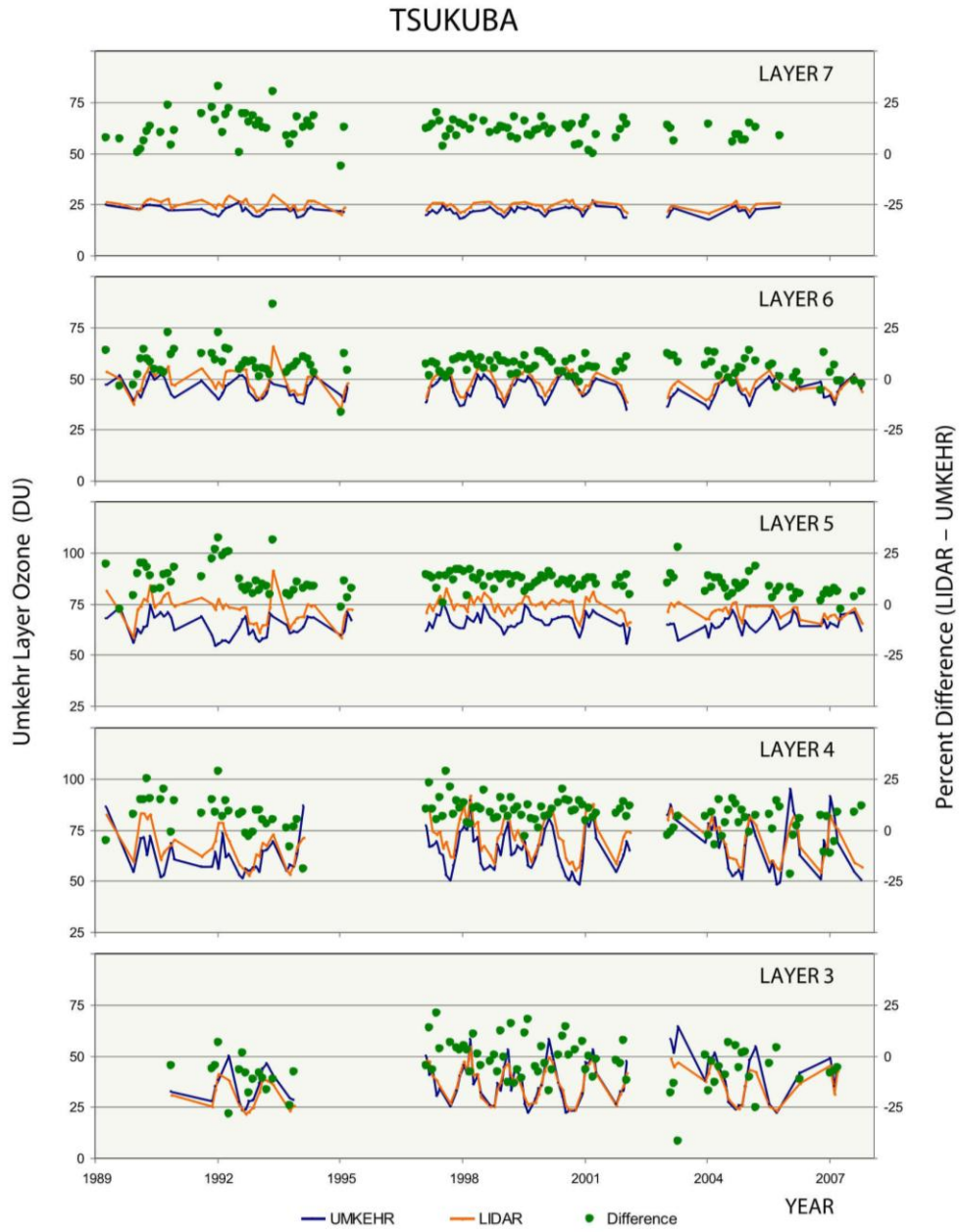


Figure 7. (continued)

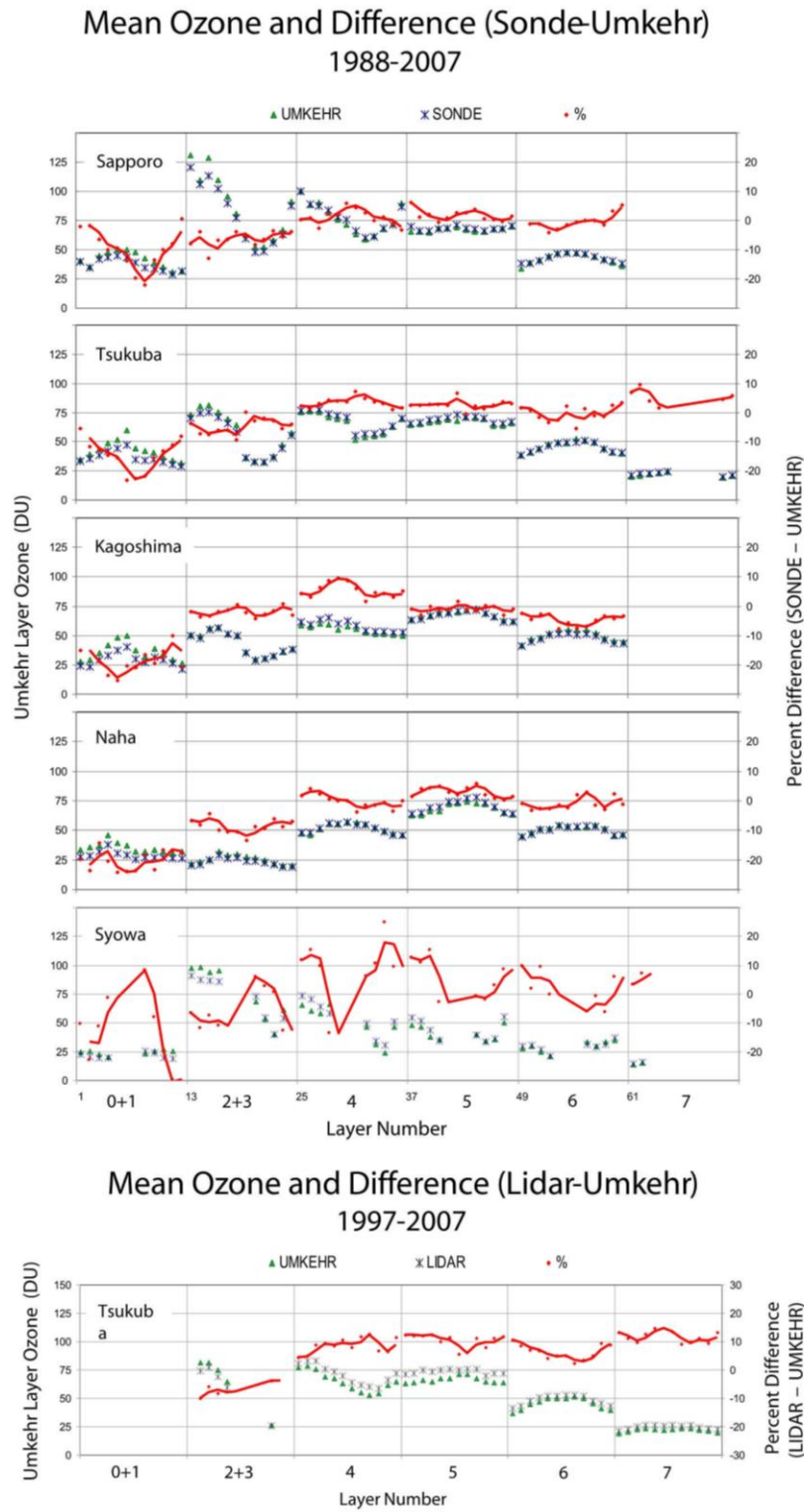


Figure 8. Mean seasonal variations of (top) ozonesonde and Umkehr ozone and their difference and (bottom) lidar and Umkehr ozone and their difference for each layer.

Table 2. Percent Difference at Each Station^a

Umkehr Layer	Sonde-Umkehr					Lidar-Umkehr Tsukuba
	Sapporo	Tsukuba	Kagoshima	Naha	Syowa	
0 + 1	-15.8 (14.6) 141	-16.4 (11.4) 192	-20.3 (13.0) 128	-19.8 (12.0) 109	-17.1 (17.0) 59	-
2 + 3	-6.9 (8.8) 141	-6.9 (7.3) 192	-3.2 (10.7) 132	-8.4 (7.0) 109	-5.3(10.9) 59	-
3	-3.7 (8.9) 141	-4.1 (7.0) 192	1.0 (10.1) 133	-6.0 (6.9) 109	0.8 (12.6) 59	-2.7 (10.4) 69
4	3.9 (9.4) 141	2.9 (6.2) 192	6.0 (7.1) 130	0.0 (6.4) 107	11.6 (16.0) 57	7.5 (7.5) 89
5	2.8 (6.6) 134	3.7 (4.9) 188	-0.2 (5.6) 123	3.0(4.9) 105	6.0 (15.3) 55	10.1 (4.5) 91
6	-0.9 (6.6) 107	1.0 (5.3) 168	-4.2 (5.6) 108	-0.7 (5.3) 92	1.5 (10.1) 43	5.8 (4.6) 86
7		4.6 (5.9) 27	8.1 (6.0) 10		5.2 (10.3) 8	11.2 (4.3) 59

^aThe statistics period of Umkehr and ozonesonde is from 1988 to 2007 (Umkehr and lidar: 1997–2007). In parentheses are shown standard deviations, followed by the number of data.

lower than Umkehr ozone probably owing to KC ozonesonde's characteristics. Quite distinct features that differ in shapes and sizes for three analyzed stations before 1990 could be caused by limited ozonesonde observation frequency (once in a month before 1990). Lidar ozone is about 5% lower than Umkehr and ozonesonde ozone in layers 4 and above after 2005, though in the years prior to 2005, lidar ozone is generally higher. Low ozone by lidar measurement around 1992 may be due to increased atmospheric aerosols from Mount Pinatubo eruption. A sudden change in characteristics of lidar data before and after 1995 may be due to an optical system change.

4.5. Comparison of Profiles

[39] Figure 10 shows the mean biases among the three methods in Tsukuba as a function of altitude (layer). Comparison periods are 1994 ~ 2007 for ozonesonde (SND) and Umkehr (UMK), 1997 ~ 2007 for lidar (LDR) and UMK, and 1997 ~ 2007 for SND and LDR, respectively. Comparison period is selected to cover the period using 125 that has large SZA dependency to show the revision effect clearly. Standard deviations are also shown. For SND and UMK comparison, Umkehr profiles retrieved from both ORG and REV are shown. The new algorithm UMK04 is used for this comparison.

[40] Comparison between SND and UMK from REV shows good consistency with differences less than 5% in layers 3 to 7. As for comparison with UMK from ORG, relatively large negative biases in upper layers 6 and 7 are noticed. Large negative biases in lower layers in both REV and ORG results are considered to be due to KC ozonesonde's characteristics, though some of it may be due to limited quality in these Umkehr layers.

[41] Comparison between UMK and LDR shows that LDR is generally higher than UMK except in layer 3, where both are almost identical. However, the offset between two systems is almost constant (from 5 ~ 10%) in layers 4 to 7. These characteristics are consistent with results reported in SAGE-II and LDR comparisons with precision less than 10% [Nakane *et al.*, 1993; Park *et al.*, 2006].

4.6. Long-Term Ozone Trend

[42] Long-term variation of ozone amount in some layers at Sapporo, Tsukuba, Naha, and Syowa retrieved by UMK04 are shown in Figure 11. Here, in order to glance at round tendency, simple linear trends are derived without removing various effects such as solar activity, QBO, etc. In Figure 11a, trends in two periods 1979–1996 and 1996–2006 for Sapporo, Tsukuba, and Naha are shown. Though the number of Umkehr

data is small before 1984 except at Tsukuba (Figure 1), it seems that the quality of linear trend of period 1979–1996 is not degraded. Detailed trend analyses considering solar activity, QBO, etc., will be done in another paper.

[43] Layer 4 is located in the most ozone abundant lower stratosphere, where the ozone decreasing tendency from the 1970s to mid-1990s is found at Sapporo and Tsukuba, and subsequent gradual increasing is found at Sapporo (Figure 11a). Layer 7 ozone accounts for only 7% of total column ozone, but is most sensitive to chlorine changes, and a decreasing trend is detected in the 1980s at three stations. Though the quality of ozone in layer 0 + 1 in the troposphere may be limited, tropospheric ozone at three stations shows monotonous increasing since the 1970s with relatively small variation. Trends and standard deviations are summarized in Table 3. The largest trend at a rate of 8.4%/decade between 1979 and 1996 is found at Tsukuba in layer 8 + 9 + 10.

[44] Ozone amount in the Austral spring (average over September through November) in layers 4 and 8 + 9 + 10 at Syowa are shown in Figure 11b. Here the trends between 1987 and 2007 are shown. In both layers, obvious decreasing trends with variations can be found.

[45] Figure 12 shows the profile of ozone trends between 1980 and 2007 at Sapporo, Tsukuba, and Naha. At Sapporo, the northernmost Japanese station, an apparent decreasing trend of about 6%/decade is observed in layers 7 and 8 in the upper stratosphere. At lower latitudes (Tsukuba and Naha) the maximum ozone decrease in the upper stratosphere is reduced and the altitude of the maximum trend is higher as compared to the northern station. In the lower stratosphere a decreasing trend can be seen at Tsukuba and Naha. However, a decreasing trend at this altitude is not detected at Sapporo, but this is due to recent increasing tendency after the mid-1990s. If we see the trend by the mid-1990s a large decreasing trend is analyzed (see Table 3).

[46] In layer 0 + 1 in the troposphere, apparent increases can be seen at Tsukuba (3.5%/decade) and Naha (5.5%/decade), though the quality of this layer's ozone may be somewhat low. An increase in tropospheric ozone is considered to be due to an increase in ozone precursor emissions. These characteristics are consistent with results reported in Scientific Assessment of Ozone Depletion 2006 [World Meteorological Organization, 2007].

5. Conclusions

[47] Umkehr measurement data are reevaluated by introducing new techniques to remove instrumental uncertainty in measured N values by assessing each instrument's character-

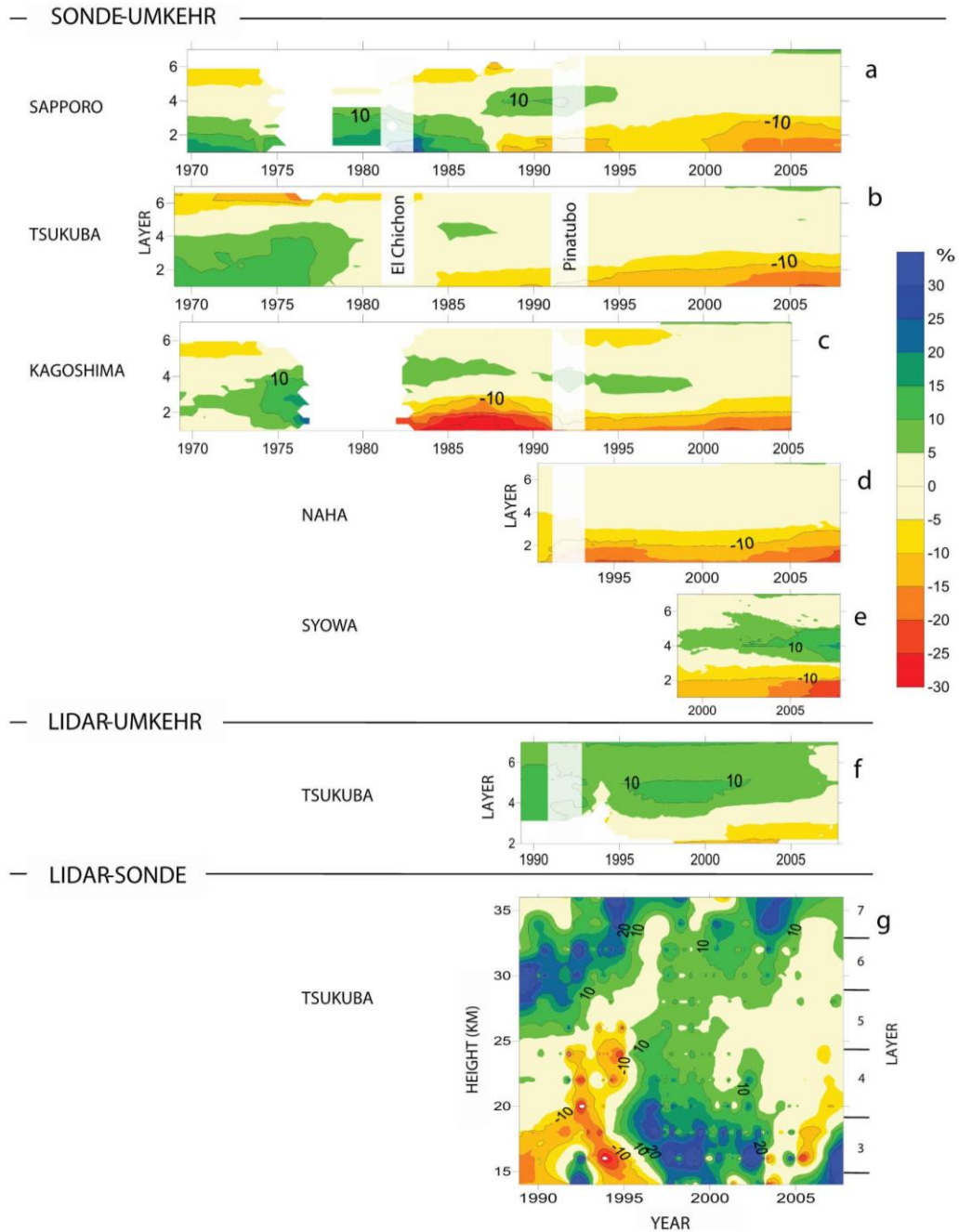


Figure 9. Vertical and temporal distribution of differences in ozone captured by three independent methods are shown. Comparisons during 3 years following the two major volcanic eruptions, such as Mount El Chichon in 1982 and Mount Pinatubo in 1991, are not shown owing to stratosphere aerosols interference in Umkehr and lidar measurements. (a–e) Differences between sonde and Umkehr data at Sapporo, Tsukuba, Kagoshima, Naha and Syowa. (f) Difference between lidar and Umkehr. (g) Difference between lidar and ozonesonde measurements at Tsukuba. Comparison is made in finer resolution in Figure 9g than Figures 9a–9f, owing to higher vertical resolution available from ozone sounding of about 100 m [Fujimoto *et al.*, 2000] and lidar measurements of about 1.5 ~ 4 km [Nagahama *et al.*, 1999]. Approximate Umkehr layers are marked on the right side of Figure 9g.

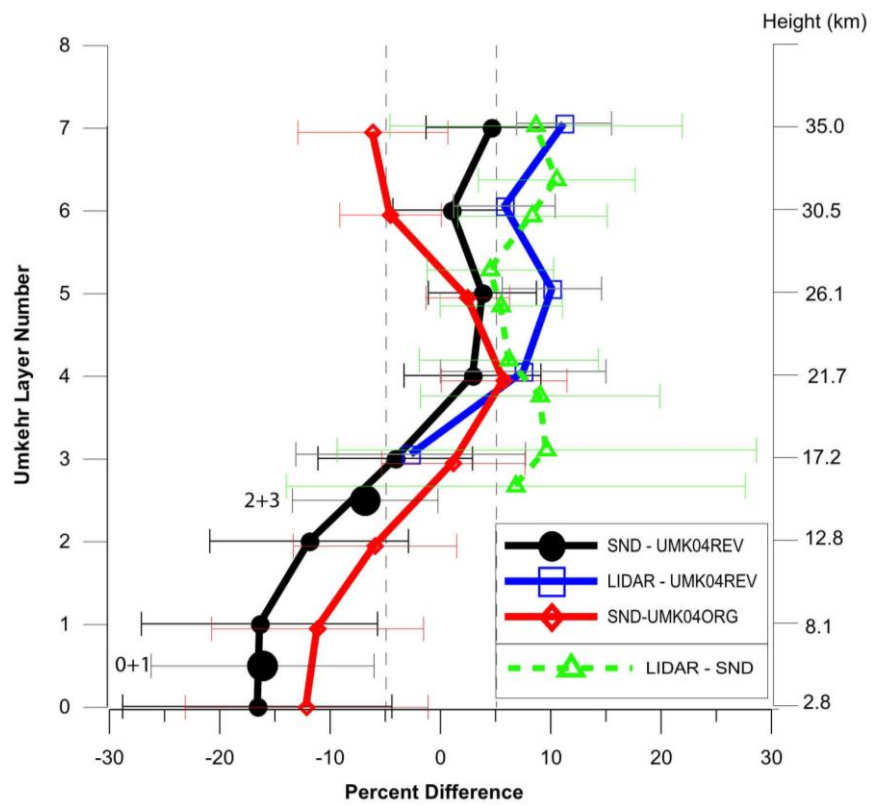


Figure 10. Vertical distribution of mean profile differences between ozonesonde and Umkehr and between lidar and Umkehr at Tsukuba are shown. As for ozonesonde and Umkehr comparison, both the vertical profile differences before and after reevaluation are shown. Statistics period is from 1994 to 2007 (1997 to 2007 for lidar and Umkehr). Error bar shows 1 standard deviation.

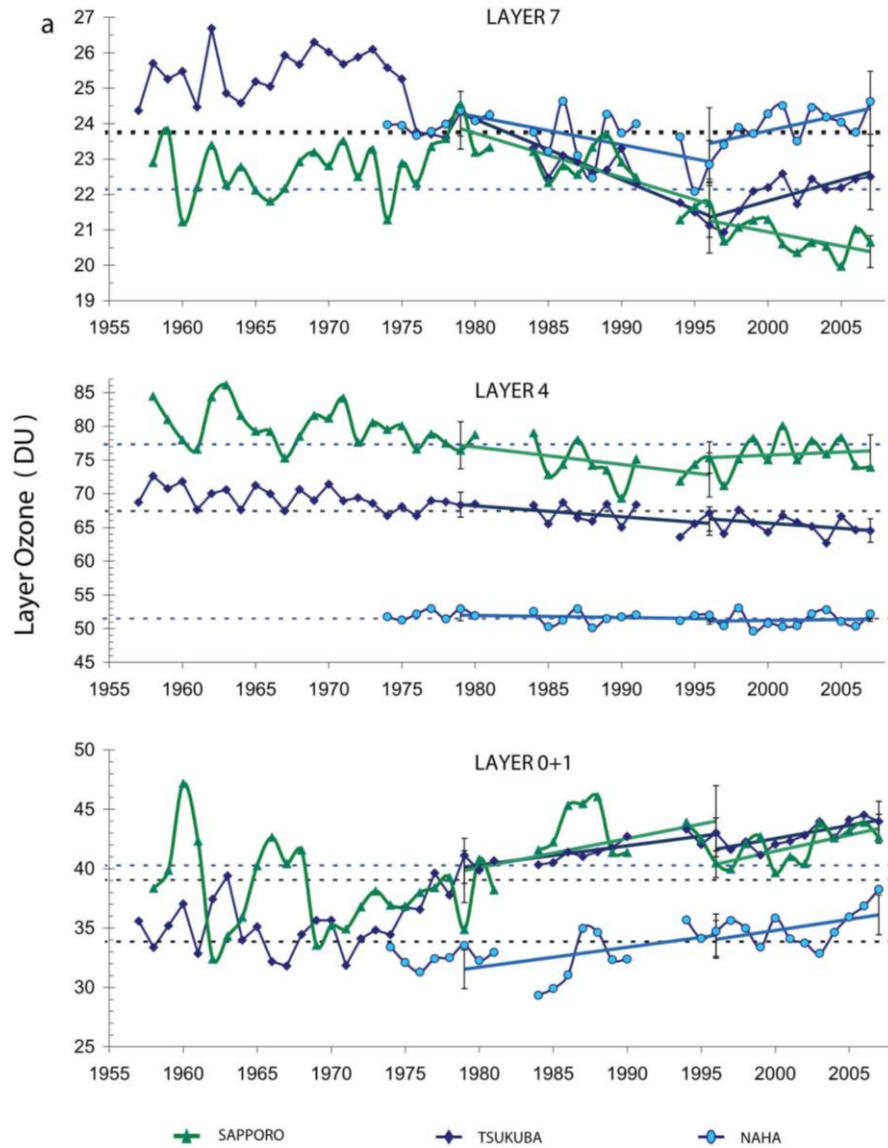


Figure 11. Long-term variation of ozone amount in three layers. Linear trend fit is derived without removing effects such as solar activity, QBO, etc. Error bar shows 1 standard deviation. (a) Annual mean ozone amount at Sapporo, Tsukuba, and Naha for layers 0 + 1, 4, and 7. Solid lines show linear trends from 1979 to 1996 and from 1996 to 2007. Dashed lines show the averages from 1960 to 2007 (from 1974 to 2007 for Naha). (b) Blue symbols and lines show monthly mean ozone amount (September through November) at Syowa in layers (left) 4 and (right) 8 + 9 + 10. Red symbols and lines show their 3-month averages, and dashed lines show their linear trend fit from 1987 to 2007.

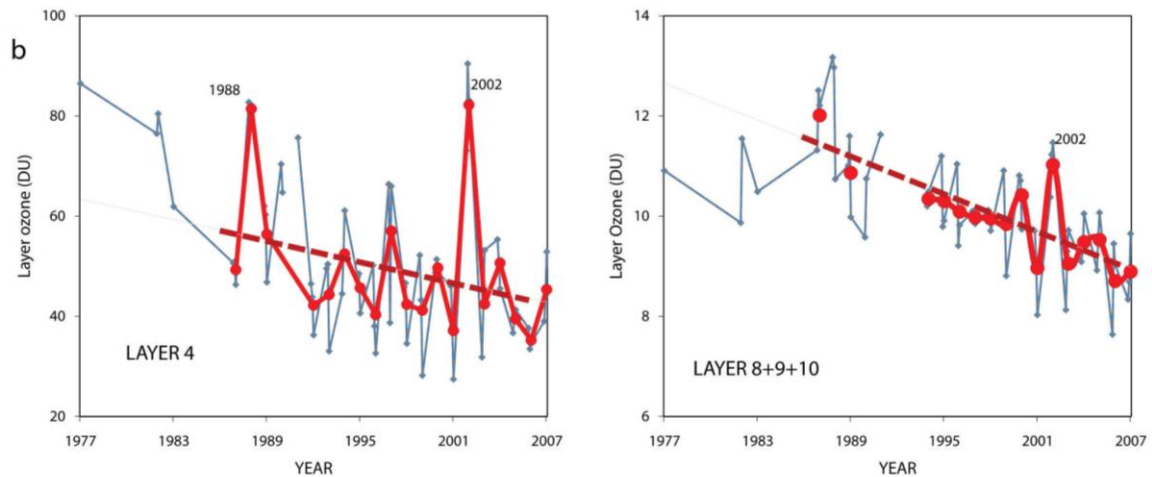


Figure 11. (continued)

istics on the basis of intercomparisons with a reference Dobson instrument. Reevaluated and original N values are processed by new Umkehr retrieving algorithm UMK04 as well as UMK92 to derive vertical ozone profiles. It is shown that the ozone profile retrieved by UMK04 shows a clear decreasing trend in the 1980s in the lower stratosphere. This trend was not analyzed when retrieved by the UMK92 algorithm, which uses a priori total ozone data that change in the long-term span. It has been noticed that the ozone profile retrieved from original N-value data shows unrealistic features in some cases. However, it is shown that this unrealistic feature is removed in the profile retrieved from revised data sets.

[48] Vertical ozone profiles retrieved from revised N-value data sets were compared with coincident ozonesonde and lidar measurements on the daily basis. The results show that the Umkehr deviation from ozonesonde profile is less than 5% in layers 4 to 6, which suggests that reevaluated Umkehr

data provide high-quality ozone profiles available for long-term trend analyses. However, more than 10% deviation in the lowermost layer is found. This is considered to be due to KC ozonesondes characteristics (though limited quality in this Umkehr layer may account for some of it), which is supported by recent collocated data comparison with the tropospheric lidar system in Tsukuba [Nakazato *et al.*, 2007].

[49] Long-term ozone trends in Umkehr measurements show the largest decreases in the upper stratosphere (layers 7 and 8 for Sapporo and Tsukuba). Decreases in the lower stratosphere (layer 4) between 1979 and 1996 are also noticed at Tsukuba and Sapporo. A decrease in total ozone in this period reflects a decrease in such a layer's ozone. Increases in 0 + 1 layer ozone (tropospheric layer) are apparent over Naha and Tsukuba, which suggests an increase in ozone precursor emission. The statistically significant results are obtained from the revised Umkehr data sets. Future analysis of the revised time series with respect to earlier studies will address

Table 3. Linear Trends and Statistics Derived for Four Stations of the Japanese Network Without Removing Effects Such as Solar Activity, QBO, etc.^a

Station	Parameter	Layer Number							
		0 + 1	2 + 3	4	5	6	7	8	8 + 9 + 10
Sapporo	Mean ozone (DU)	42.3	86.6	74.1	66.6	44.2	22.8	9.2	13.7
	%/decade	6.6	1.9	-3.8	-5.8	-6.3	-5.6	-3.4	-1.8
	SD toward trend (%)	6.8	5.7	3.1	3.2	3.4	2.4	4.0	3.2
	95% confidence (%)	3.6	3.0	1.6	1.7	1.8	1.3	2.1	1.7
Tsukuba	Mean ozone (DU)	41.7	58.0	66.7	67.1	45.4	22.7	8.9	13.2
	%/decade	5.7	-0.1	-3.7	-3.0	-2.4	-7.2	-11.4	-8.4
	SD toward trend (%)	3.0	0.9	2.7	1.8	1.3	1.6	2.7	2.1
	95% confidence (%)	1.6	0.5	1.4	0.9	0.7	0.8	1.4	1.1
Naha	Mean ozone (DU)	33.1	24.5	51.7	69.8	50.1	23.4	8.5	12.6
	%/decade	5.1	0.3	-0.4	-2.2	-2.0	-3.4	-5.9	-4.3
	SD toward trend (%)	5.2	0.4	1.6	1.7	2.2	2.6	3.7	3.1
	95% confidence (%)	2.7	0.7	0.8	0.9	1.2	1.4	1.9	1.7
Syowa	Mean ozone	24.7	50.9	35.3	40.9	33.5	16.6	6.6	10.1
	%/decade	-4.5	-14.3	-24.0	-8.9	-1.9	-5.7	-10.5	-7.4
	SD toward trend (%)	6.9	23.1	31.5	16.6	5.9	5.1	6.1	4.3
	95% confidence (%)	3.2	10.7	14.5	7.7	2.7	2.4	2.8	2.0

^aTrends are given in %/decade. Analyzed periods are 1979–1996 for Sapporo, Tsukuba, and Naha and spring months (September, October, and November) of 1987–2007 for Syowa. Periods interfered with by volcanic eruptions, El Chichon (1982–1983) and Mount Pinatubo (1992–1993), were removed from the analysis.

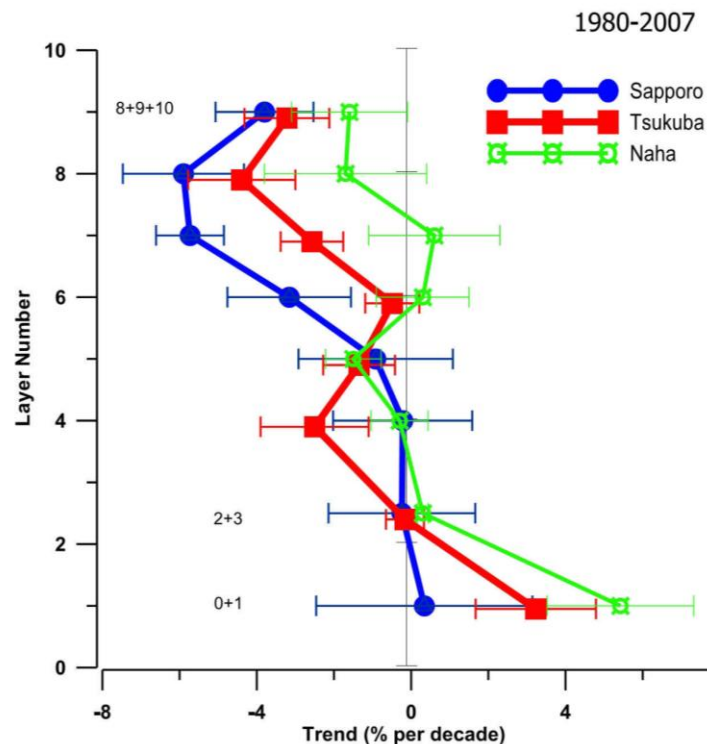


Figure 12. Profile of ozone trends between 1980 and 2007 at Sapporo, Tsukuba, and Naha. Error bars show the 95% confidence limits.

the long-term and short-term variability and the effects of the applied corrections. This new data set is now made available for interested researchers from the WOUDC.

[50] Calibration method and reference standard instrument for Umkehr measurements have not been established yet. Results of this study show that the unified intercomparison procedure of Umkehr measurements introduced here with the fixed reference standard (such as 083 and 116) can provide the international Dobson network with a great improvement of data quality.

[51] **Acknowledgments.** We would like to thank to the staff of WOUDC, NDACC, NOAA, NIES, and JMA for their kind support in immediate data usage. Also, we would like to thank members of the WMO/Ozone SAG meeting for discussion regarding the analysis of this study.

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Vertical Ozone Profile by Umkehr Measurements at Syowa Station

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Vertical ozone profile by Umkehr measurements at Syowa Station

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Dobson Umkehr measurement for derivation of the vertical ozone profile has been conducted at Syowa station (69.0°S, 39.6°E) in Antarctica since 1977. Introduction of a highly automated measuring system in 1994 at Syowa has brought about many opportunities and high quality data acquisition for Umkehr measurements. Short Umkehr measurement (A, C and D pairs) has been routinely conducted at Syowa, where solar zenith angle is generally high, except for the polar summer when the solar zenith angle is too low. In this study, recently re-evaluated N-value records acquired at Syowa and the new algorithm UMK04 are used to deduce ozone profiles. The ozone hole in 2006 was developed to the largest size ever observed, and at Syowa the lowest total ozone of 114 DU since the beginning of observation was recorded. Umkehr measurements also show extremely low ozone amount in each layer above Syowa. This is the minimum record in Umkehr data at Syowa since the beginning of observation in 1977. The ozone profiles for recent years show decreasing tendency in spring in the upper (about 40 km) and lower stratosphere.

1. Introduction

Total ozone observations using the Dobson spectrophotometer have been routinely conducted at Syowa Station in Antarctica since 1961. An abrupt ozone decrease observed in the Austral spring in 1982 at Syowa (Chubachi 1984) as well as the decreasing trend since the 1970s at Halley Bay (Farman *et al.* 1985) led to the discovery of the ozone hole, which led to the Montreal Protocol on Substances that Deplete the Ozone Layer and its following amendments. The Antarctic stratosphere is the most sensitive region to global ozone depletion, so it is important to see the situation of Antarctic ozone (not only total ozone but profile ozone) to confirm the effects of regulation by the Montreal Protocol and to make future policy decisions.

Vertical ozone observations by the Umkehr method at Syowa started in 1977, and have been almost routinely conducted since 1987. At Syowa Station, which is located at high latitudes, Umkehr measurements taken at sun elevations between 60° (or 70°) and 90° solar zenith angles (SZA) are possible in the period between late September and middle of March. Ozone profiles that are routinely retrieved from Umkehr measurements (Petropavlovskikh *et al.* 2005) provide an adequate dataset for assessment of trends in stratospheric ozone and for validation of satellite observations. Syowa is one of the most important stations that can provide such data from Antarctica.

In 2006 the ozone hole had developed to the largest size ever observed, and at Syowa the lowest total ozone since the beginning of observation was recorded. This is

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due to the fact that the concentration of ozone depleting substances in the stratosphere is still at a relatively high level (World Meteorological Organization (WMO) 2007). For this study, datasets re-evaluated using the method of Miyagawa and Hirose (2004) and Miyagawa *et al.* (2009) were used in order to remove instrumental errors in Umkehr records. These long-term datasets were processed by Umkehr retrieving algorithm UMK04, which was recently revised by Petropavlovskikh *et al.* (2005) to deduce vertical ozone profiles. Retrieved ozone profiles reveal developing features in each layer at the time of the 2006 ozone hole. The long-term trend in the stratosphere over Syowa Station is also discussed.

2. Data

About 1240 data records that were acquired by the Japan Meteorological Agency (JMA) at Syowa Station between 1977 and 2007 are used in this study (see table 1). All these data are available at the World Ozone and UV Data Centre (WOUDC). In addition to C-pair, the measurements at the Dobson A- and D-pairs have been acquired at Syowa since the fully automated operating system was installed in December 1994. The number of annual measurements increased significantly after the introduction of the automation system, and 94 measurements were done during the year 2006. N-value data at SZA 70° through 90° are used in derivation of ozone profiles by the UMK04 algorithm. The JMA's global analyses dataset is used for conversion of altitude unit of vertical ozone profiles.

Ozone concentration by KC ozonesonde is deduced by the newly revised algorithm that resolved some problems found in the Julich OzoneSonde Intercomparison Experiment (Fujimoto *et al.* 2000). The improvement of the sounding data when processed by the revised algorithm was confirmed in the Balloon Experiment on Standards for Ozonesondes (Deshler *et al.* 2008).

3. Results and discussion

In 2006 the ozone hole developed to the largest size ever observed, and the minimum record of total ozone since the beginning of observation in 1961 (114

Table 1. Number of obtained ozone profiles using C-pair data and months of measurements at Syowa. The columns # Obs and # Mon show the number of observations and months, respectively, in which ozone profiles are obtained in each year.

Year	# Obs	# Mon	Year	#Obs	#Mon
1977	3	2	1996	61	8
1982	14	4	1997	60	7
1983	2	1	1998	39	9
1987	23	4	1999	50	9
1988	42	8	2000	62	7
1989	88	9	2001	58	8
1990	24	6	2002	89	8
1991	22	6	2003	71	8
1992	31	7	2004	101	9
1993	49	8	2005	77	8
1994	58	8	2006	88	7
1995	56	9	2007	72	7

DU on 17 October 2006) was observed at Syowa. Ozone profiles by Umkehr and ozonesonde measurements in 2006 show low concentrations, mainly at altitudes between 10 and 25 km during the ozone hole period (figure 1). The number of collected ozone profiles is 59 by Umkehr and 18 by ozonesondes between September and November 2006. The profiles retrieved by the UMK04 model on 27 September and 15 October 2006 in layers 4 and 3, respectively, show 0 DU ozone. This value is the minimum record since the beginning of observation in 1977. The value on 15 October 2006 is consistent with the data recorded by ozonesonde measurement at 10 a.m. (local time) on the same day. Satellite observation shows that ozone hole area (total ozone less than 220 DU) covers Syowa at this period of time. The typical ozone amount in layers 3 and 4 is about 10% of the ozone amount observed in summer time.

Figure 2 shows the seasonal and long-term ozone variation in each layer by Umkehr measurements from 1977 to 2007. In 2006, the ozone amount remained at typical ozone levels in the pre-ozone hole season (January to April) and plunged to the lowest level ever observed in spring (September to November) in all layers except in layer 1 (figure 2a). Tropospheric ozone in layer 1 was observed at the typical level, but dropped to rather lower levels in November. The ozone amount in layers 3 and 4 was extremely low, especially around 1 October. Long-term ozone data are separated into two sets with regards to the total ozone level: larger than or less than 220 DU. These two sets of data are plotted separately in figure 2b. The average ozone amount in layers 2 to 5 in the sub-set representing ozone hole conditions (total ozone less than 220 DU) is 60–30% lower than averaged ozone amount calculated for non-ozone hole conditions (larger than 220 DU). Ozone amounts for inside/outside ozone hole conditions in other layers are roughly at the same level. This suggests that ozone depletion in the ozone hole occurs mainly in the lower stratosphere (centred in layers 3 and 4).

Figure 3 shows the long-term ozone trend in layers 8 and 9 (the upper stratosphere) and 4 (the lower stratosphere) for spring and summer. The time series of monthly mean deviations from the averages are plotted for the 1977–2007 time period for spring and summer seasons (spring is September to November and summer is December to March). In this analysis, components of natural ozone variations related to solar activity and QBO are removed. Ozone in the spring season shows clear and steady decreasing both in the upper (9.6%/decade) and the lower (16.6%/decade) stratosphere. Conversely, ozone in the summer shows only a small decrease in the upper stratosphere and no clear decrease in the lower stratosphere. Table 2 shows the linear trend for each layer. Ozone decrease is found in two altitude regions though it is insignificant in the summer lower stratosphere.

Sudden ozone increase is found in spring of 2002 both in the upper and lower stratosphere. This is due to the major stratospheric sudden warming that suppressed ozone depletion in that year (Varotsos 2002, 2003, 2004). Another sudden ozone increase synchronized with stratospheric sudden warming is found in the lower stratosphere in 1988. However, the ozone amount in spring of 2006 shows the minimum record values as discussed before. This may be related to the minimum record values observed in the upper stratosphere in the summer of 2007.

Figure 4 shows the progress in ozone amount in layer 4 in the lower stratosphere in 2006 and other years. Ozone in 2006 decreases from August to early October then increases thereafter. Ozone loss rate in the initial phase of 2006 is 0.046 ppmv/day,

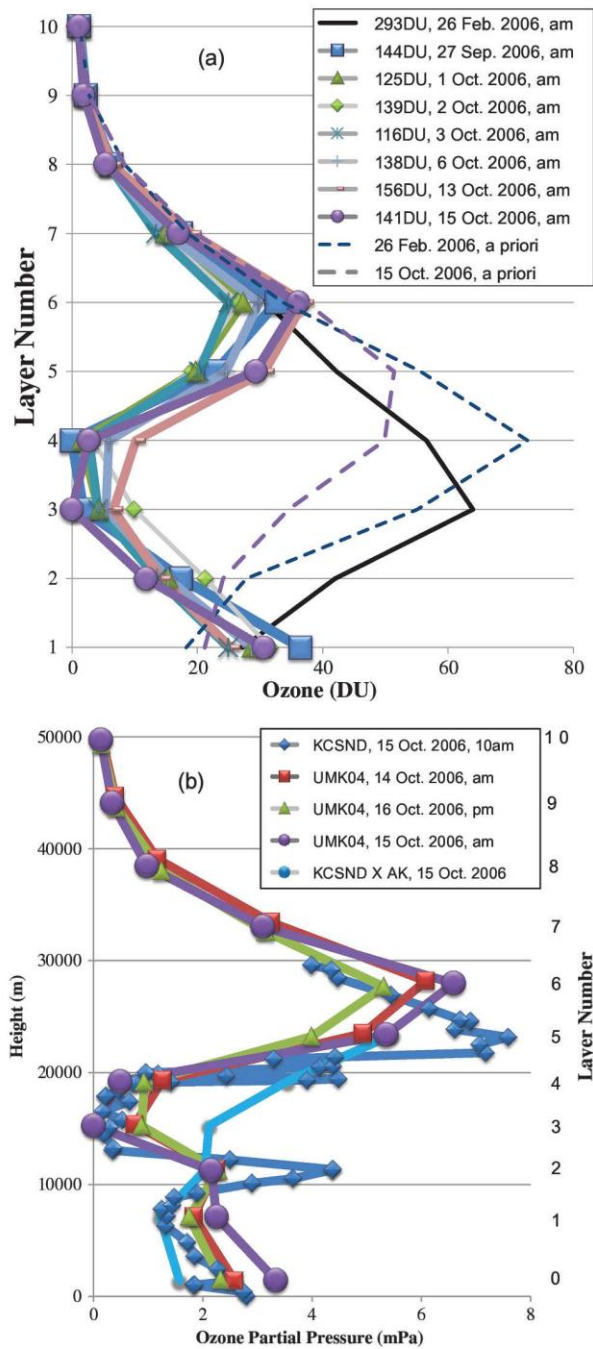


Figure 1. Umkehr ozone profiles in the ozone hole season in 2006. (a) Profiles from 27 September to 15 October. The dashed line shows *a priori* profiles used in retrieving by UMK04 algorithm. (b) Selected Umkehr profiles and ozonesonde profile around 15 October when the minimum ozone amount in layer 3 is observed.

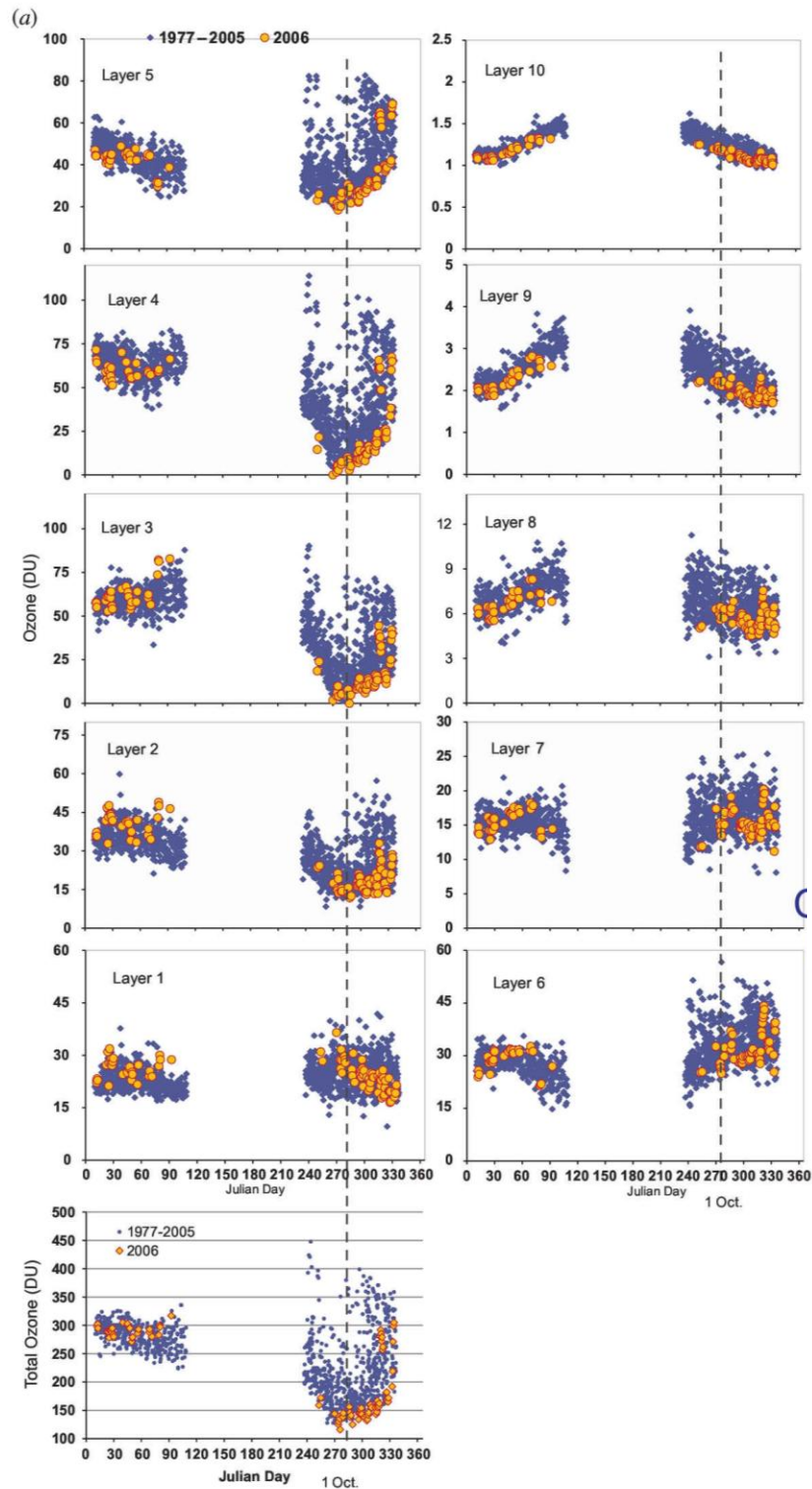


Figure 2. Ozone amount in each layer by Umkehr measurement shows seasonal and long-term variation from 1977 to 2007. Total ozone variation is also shown in the bottom line. (a) The seasonal ozone variation in 2006 and 1977–2007 for each layer. A dashed line shows the day when total ozone is the lowest of the year (1 October 2006). (b) The long-term ozone variation for each layer. According to total ozone, all data are sorted by red dots (less than 220) and blue dots (over 220 DU). The dashed line shows linear fit.

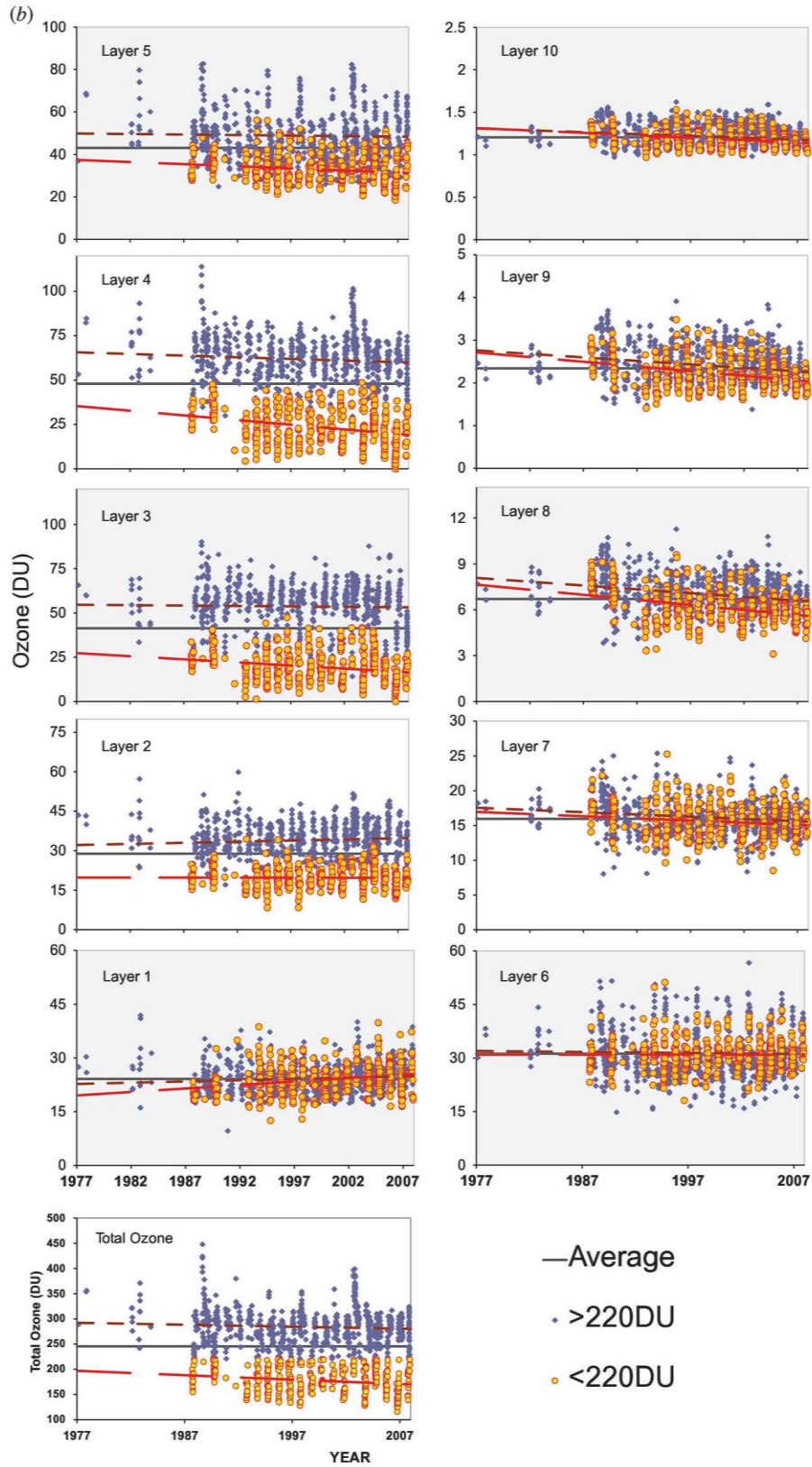


Figure 2. (Continued).

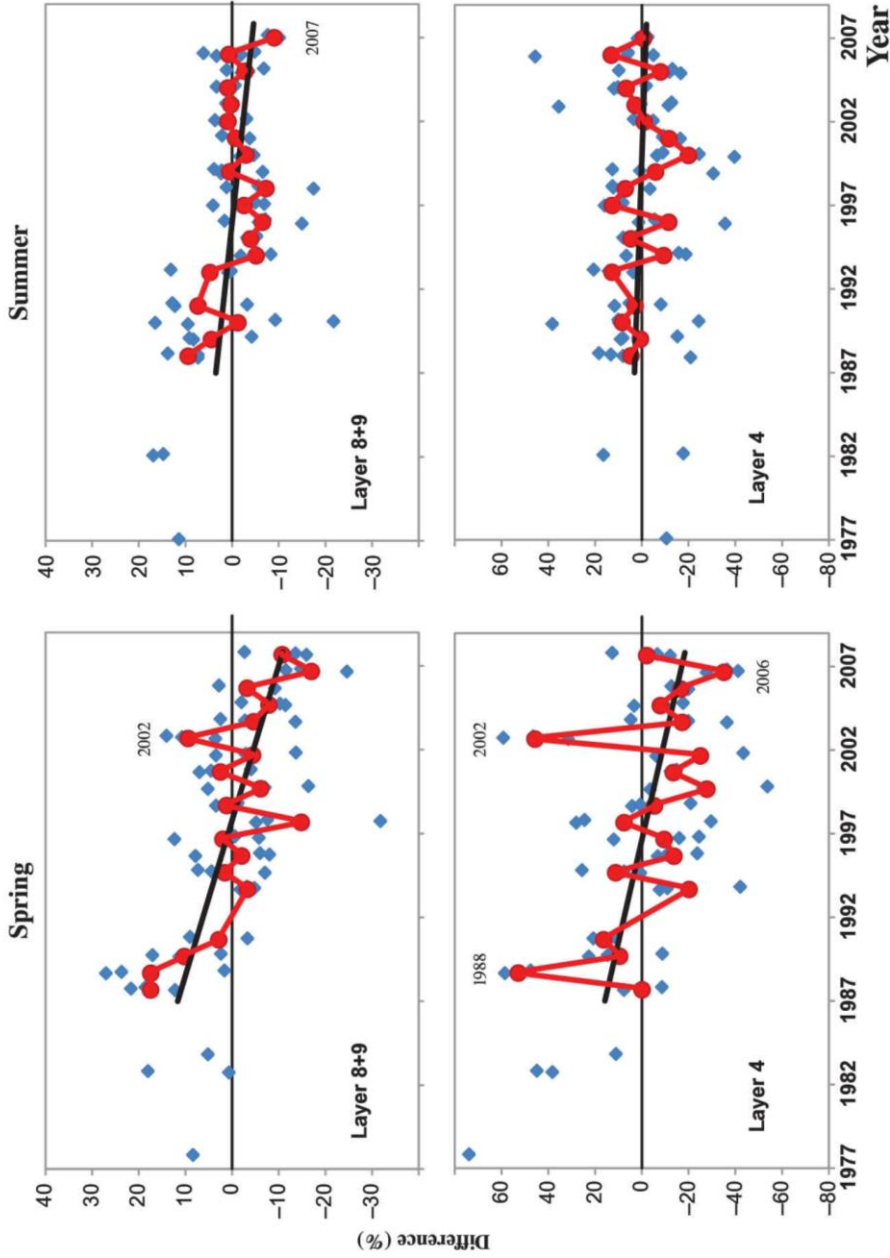


Figure 3. Long-term variation of ozone amount in layers 8 + 9 and 4 for spring and summer at Syowa. The blue squares shows the monthly deviations (September to November for spring and December to March for summer) from the average over 1977–2007, and red circles shows seasonal averages. In the analysis, effects of solar activity and QBO are removed from the data from 1977 to 2007. The thick line shows the linear trend fit from 1987 to 2007.

Table 2. Linear trend from 1987 to 2007 for spring (September to November) and summer (December to March) time periods in each Umkehr layer at Syowa. In the analysis, effects of solar activity and QBO are removed from the data from 1977 to 2007.

Layer	% / decade		95% confidence, %	
	Spring	Summer	Spring	Summer
1	5.2	3.9	2.8	2.1
2	-5.9	6.6	4.5	2.9
3	-13.9	2.8	5.8	3.2
4	-16.6	-2.4	6.7	3.8
5	-9.1	-2.1	4.9	2.6
6	-0.9	-0.7	2.9	1.8
7	-5.4	-2.8	2.8	1.8
8	-10.0	-4.0	3.1	1.9
2 + 3	-10.7	4.3	5.2	2.9
8 + 9	-9.6	-3.7	2.9	1.8

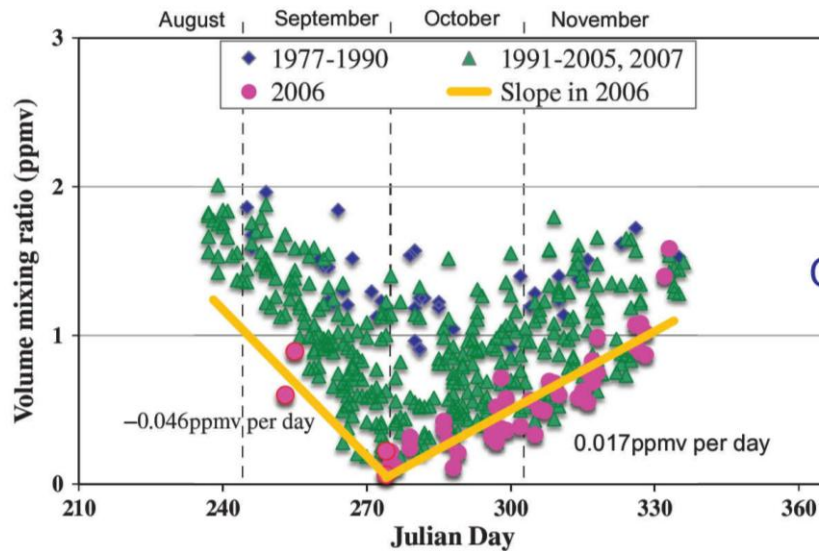


Figure 4. Time series of ozone amount in layer 4. Pink, green and blue symbols show data for 2006, 1991–2007 and 1977–1990, respectively. Yellow lines show approximate ozone changes in the decreasing and recovering phases in 2006.

which is larger than the loss rate of 0.03 ppmv/day in the Arctic region (Hirota *et al.* 2005). Return rate in the following phase is 0.017 ppmv/day, about one third of the loss rate in early spring.

4. Conclusions

Recent studies (Miyagawa *et al.* 2009) lead to re-evaluation of JMA Umkehr datasets. The revised time series of ozone profiles retrieved by UMK04 algorithm are available (WOUDC archive). Based on these studies it is expected that more accurate and stable ozone profiles can be obtained to assess long term trends. Figure 5 shows the time series of daily comparison between Umkehr and ozonesonde measurements for layer 6. Here, the ozonesonde profile is smoothed by applying the averaging kernel of the

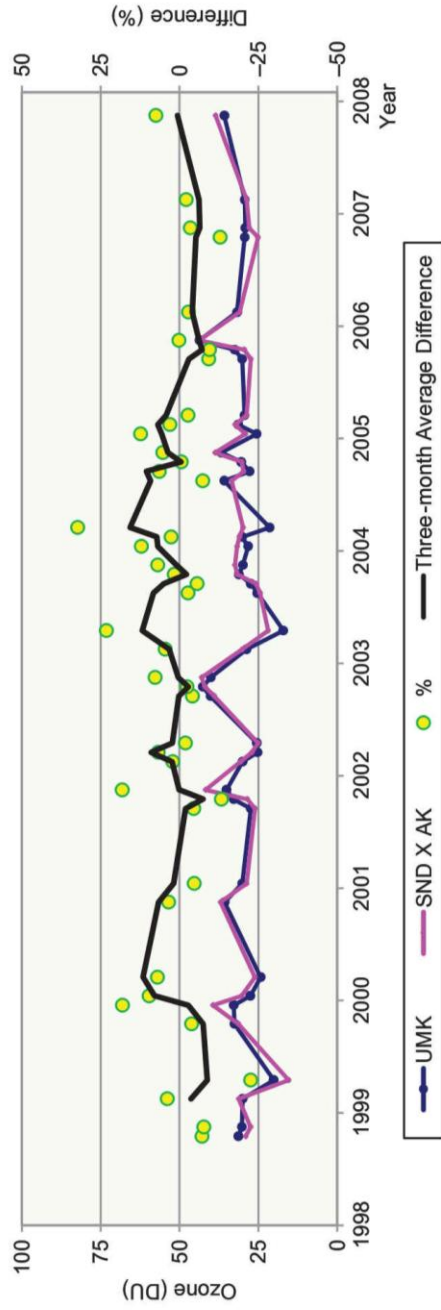


Figure 5. Time series of daily ozone comparison between Umkehr (thin blue line) and ozonesonde (thin pink line), and their difference (yellow circle) and its three-point averages (thick black line) in layer 6.

Umkehr algorithm. Both measurements show fairly good consistence with each other (relative error of about 1.5%, i.e. standard error of 10%), including seasonal variation. Umkehr data are expected to provide important information about stratospheric ozone trends in the Antarctic region in addition to the satellite data. The independent data source from Umkehr measurements is also expected to validate satellite observations. Re-evaluated new Umkehr datasets contribute to the ozone layer assessment with regards to the Montreal Protocol success.

The ozone hole in 2006 was developed to the largest size ever observed, due to the still high concentration of ozone depleting substances in the stratosphere (Reid *et al.* 1998, Schulz *et al.* 2001). By Umkehr measurements, detailed stratospheric ozone changes in the developing and recovering phase of the ozone hole are revealed.

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Chapter 3

Long-term Ozone Trends in Umkehr Measurements at Japanese stations

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LONG-TERM OZONE TRENDS IN UMKEHR MEASUREMENTS AT JAPANESE STATIONS

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Abstract

Umkehr observations have been made routinely at Japanese stations at Sapporo, Tsukuba, and Naha, and at the Antarctic station, Syowa, for more than 50 years. The discontinuous gaps in Japanese Umkehr data record have been associated with instrument replacements. Therefore, N-value data were recently reevaluated based on instrument intercomparisons. The data analysis revealed systematic errors that depend on solar zenith angle, total ozone, and other instrumental factors. The UMK04 ozone profile retrieval algorithm is applied in the processing of all reevaluated N-value time-series. We present a long-term ozone trend determined from the newly re-processed Umkehr ozone profiles.

The long-term trend in upper stratospheric ozone is discussed in this paper. Long-term variations of the ozone amount derived by UMK04 algorithm in the combined 8 and 9 layers at Sapporo, Tsukuba, Naha and Syowa are shown in the Figure 1. Linear trends in two separate periods 1970 (or 1977)-1996 and 1996-2008 are also shown for each station. Trend analyses suggest a significant decrease in the upper stratosphere over Japan during 1980s. The upper stratospheric ozone levels at Tsukuba station have shown a steady increase at 5%/decade rate after 1996. At the same time, a 7.7%/decade decrease in ozone is found in Umkehr data taken at Sapporo station, which indicates an even stronger ozone depleting rates as compared to ozone depletion rates prior to 1996. Over the Antarctic station Syowa, upper stratospheric ozone has been at a low level since 1990s. Especially low values can be seen in the last few years. Observed difference in the upper stratospheric ozone changes may be reflecting the latitude dependence of ozone depletion.

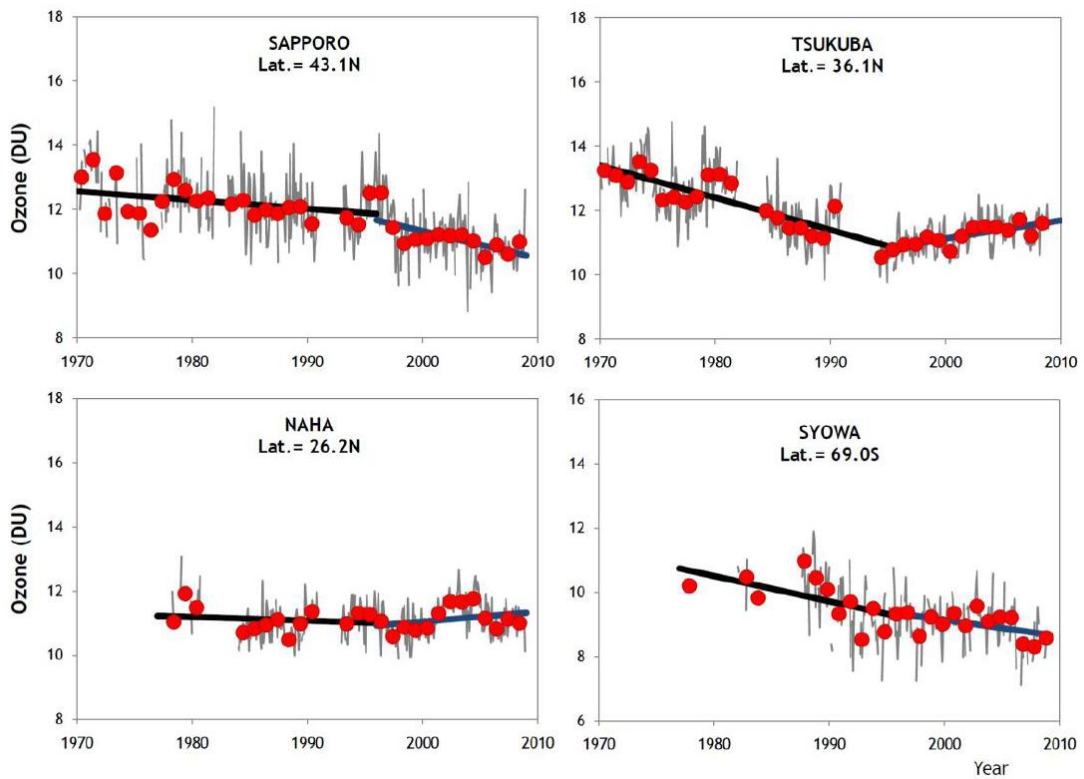


Figure 1. Long-term variations of ozone amount in 8 and 9 layers.

Linear trend fit is derived after removing effects such as solar activity, QBO, and atmosphere turbidity. A gray line shows monthly average and red circle shows average of the year. Solid lines show linear trends from 1970 (or 1977) to 1996 and from 1996 to 2008. Periods interfered by volcanic eruptions, El Chichon (1982-1983) and Mt. Pinatubo (1992-1993), were removed from the analysis.

1. Long-term variations

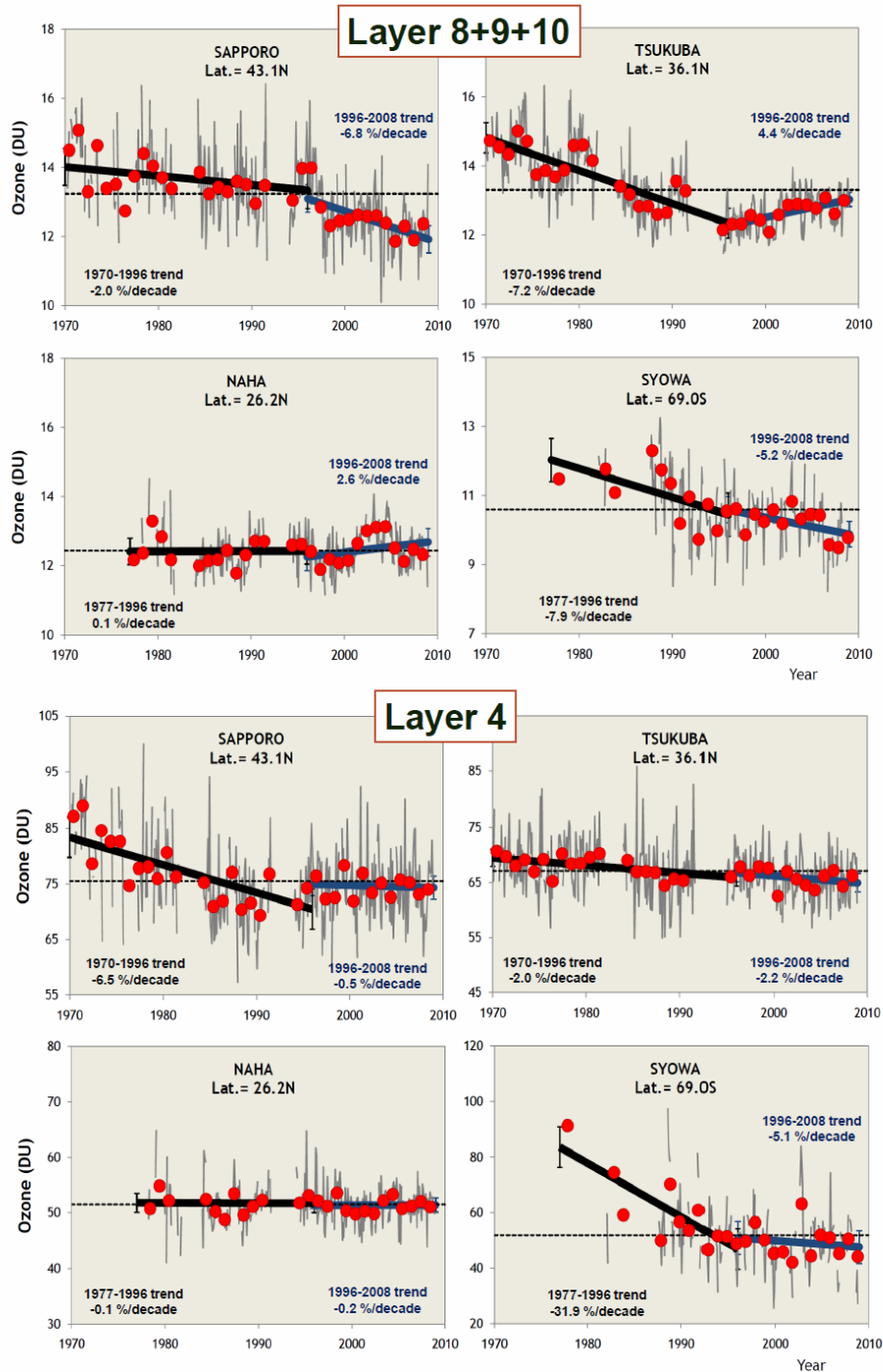


Figure 1a. Long-term variations of ozone amount in layer 8+9+10, and layer 4. Linear trend fit is derived after removing effects such as the seasonal variation, solar activity, QBO and atmospheric turbidity. A gray line shows monthly average and red circle shows average of the year. Solid lines show linear trends from 1970 (or 1977) to 1996 and from 1996 to 2008. Periods interfered by volcanic eruptions, El Chichon (1982-1983) and Mt. Pinatubo (1992-1993), were removed from the analysis.

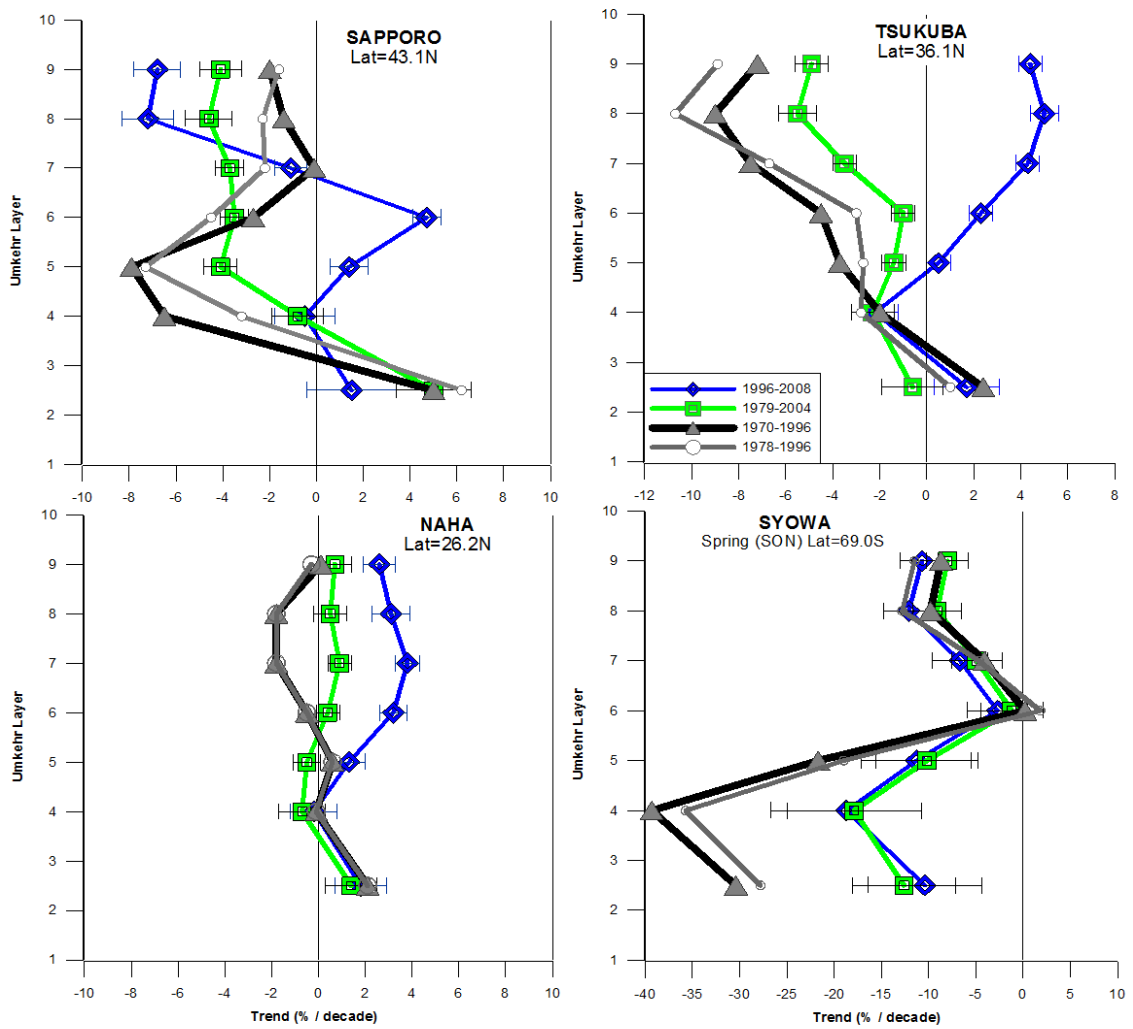


Figure 2. Ozone trends (% per decade) for four period statistics at Sapporo, Tsukuba, Naha, and Syowa. Error bars show the 95% confidence limits.

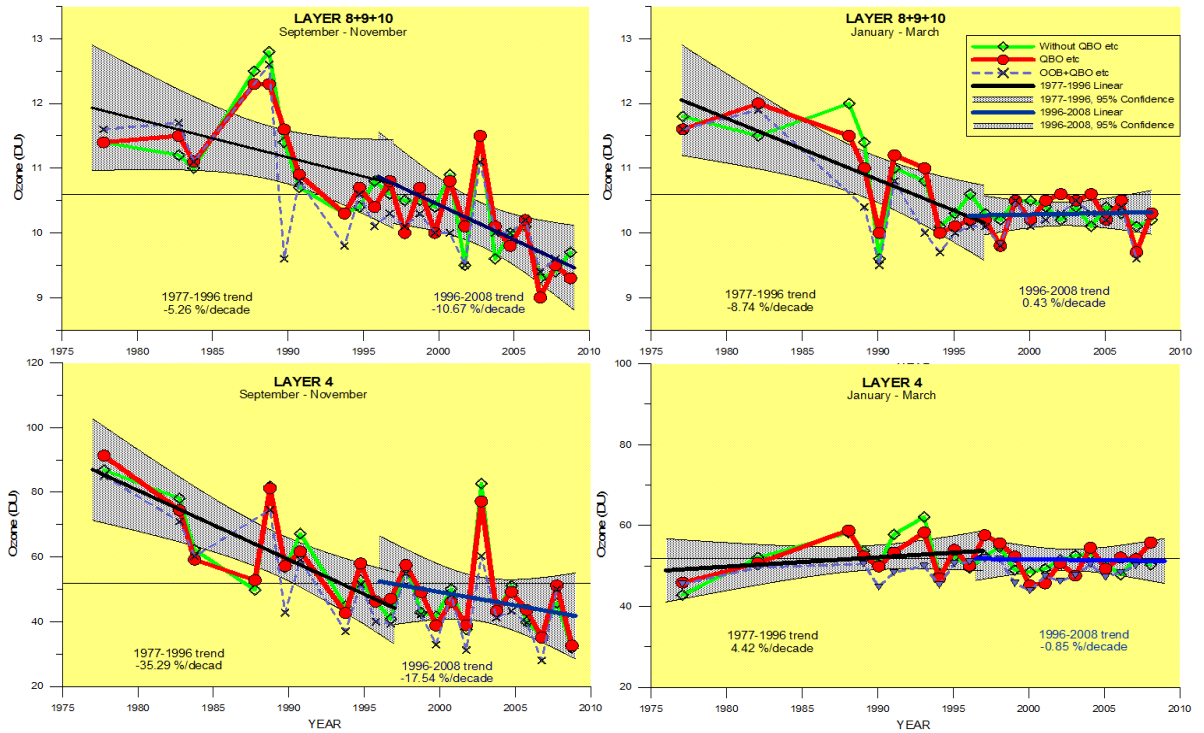


Figure 3. Same as Figure 1a, but for spring and summer time data at Syowa.

A red line shows a result removing seasonal variation, solar activity, QBO and atmospheric turbidity, and a green line shows a result of removing only seasonal variation. A dashed line shows a result including stray-light correction (OOB) in measurement data. Shaded area shows 95% confidence limit and a value shows a linear trend after removing all variation components such as QBO.

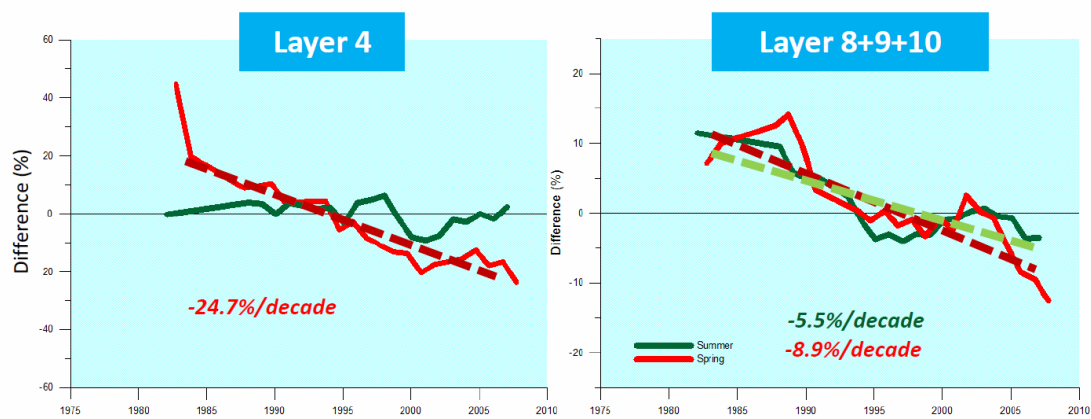


Figure 4. Same as Figure 3, but for the three-year running average for spring and summer time.

In the analysis of layer 4, data of 2002 are removed, when the stratospheric temperature was high and remarkable Antarctic ozone depletion was not observed.

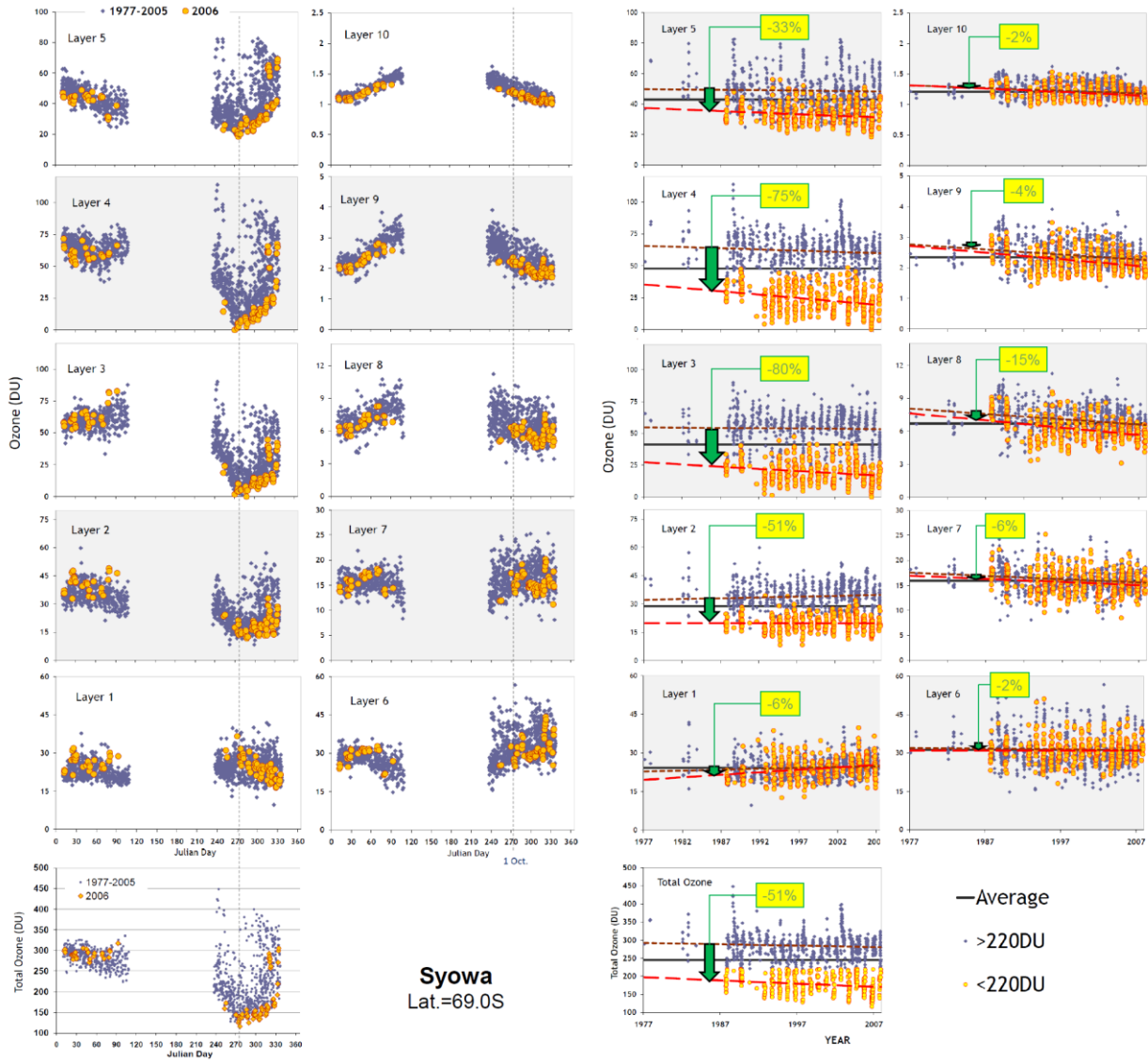


Figure 5. Seasonal and long-term variations of ozone amount in each layer by Umkehr measurement are shown from 1977 to 2007.

Total ozone variation is also shown in the bottom line. Left: The seasonal ozone variation in 2006 and 1977-2007 for each layer is shown. A dashed line shows the day when total ozone is the lowest of the year 2006 (1 October). Right: The long-term ozone variation sorted by total ozone (red dots for less than 220 and blue dots for more than 220DU) are shown. The value in figure shows a difference in decreasing ratio of two cases.

2. Discussion and Conclusion

In the upper stratosphere over Japan, the long-term ozone depletion is seen in higher latitudes (Sapporo and Tsukuba). Decreasing rate at Sapporo is even stronger (-6.8%/decade) after 1997 than before. At Tsukuba, a trend turned positive (4.4%/decade) after a long-term decreasing in the period 1970-1996 (Fig.1 and 3). Trends by SBUV/2 for 1979-2004 are generally consistent with that by Umkehr data over Japan (Fig. 2 and 3).

At Syowa in the Antarctica, a linear trend became flattened in the period 1996-2005 after a decreasing (-7.9%/decade) in 1977-1996, though recent data show a decreasing trend again. Very low ozone in the higher latitude upper stratosphere (Sapporo and Syowa) is noticeable in recent years. In the lower stratosphere, clear ozone decreases were found at Sapporo and Syowa (-6.5, -31.9%/decade, respectively) from 1970s to 1996, however, trends became almost flattened after that. These features correspond to the changes in total ozone. The trend analyses in the upper and lower stratosphere in spring and summer at Syowa show that a decreasing trend is generally found except in the lower stratosphere (layer 4) in summer. Especially a large ozone decrease is found in last 3 years (2006-2008) in layer 8+9+10 in spring, which lasted in the following summer. The 2006 was the “unusual” year, when extremely low ozone concentration was observed in most layers (except for layer 1) accompanied with low temperature. Total ozone above and below 220 DU generally corresponds outside and inside of the polar vortex, respectively. The difference in trends between outside and the inside of the polar vortex is large in the layers 2-5 (the lower stratosphere) and 7-9 (the upper stratosphere). It is said that ozone depletion in the upper stratosphere is caused by only gas phase chemistry while in the lower stratosphere it is driven by heterogeneous chemistry. The Umkehr observation provides useful data for these studies.

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Supplement: Ozone and temperature changes in the upper stratosphere

In this paper, we considered the ozone and temperature changes in the upper stratosphere using the re-analysis station data of NCEP/NCAR (National Centers for Environmental Prediction / National Center for Atmospheric Research) and JRA-25 (Japanese Re-Analysis 25 years). The main characteristics from 1982 to 2008 are as follows. For the ozone record analysis, seasonal variation, effects of solar activity, QBO and aerosol, etc. signals are removed from the data.

1. Temperature of the upper stratosphere and lower stratosphere

Figure 1 shows seasonal change of the stratosphere temperature for two specific years: 2002, when the extend of ozone destruction has receded, and 2006, when the record-breaking depth and area of ozone hole loss was recorded in the Antarctica. In the late spring (October and November) when the solar elevation angle in the Antarctic region is higher, variation in ozone and monthly mean temperature of 100 hPa and 5 hPa show high negative correlation (see Figure 2). This relation is seen in the Antarctic region and is not obvious in the Umkehr ozone record at Lauder, NZ station located at mid-latitudes in the Southern hemisphere. This late spring time period in Antarctica features the highest temperature in the upper stratosphere.

2. Relation between ozone and temperature

Figure 3 shows the annual change of layer 8 ozone of Syowa and 5hPa temperature at several Southern high latitude stations. Long term records of ozone and temperature monthly averages measured in November show high correlation where a decline of temperature and the increase in ozone is observed. The scatter diagrams and linear trends for ten years (1999-2008) are shown in Figure 4. The ozone in Syowa shows high correlation (correlation coefficient is 0.94), as do other stations in the Antarctic region. Slope at the three stations is -0.135DU per degree C.

The above analyses suggest the temperature dependence of ozone change in the upper stratosphere at 40 km altitude is about 2% per 1 degree C.

3. Conclusions

The monthly averages of the ozone (layer 8) and temperature (at 5 hPa level atmospheric pressure) for the month of November show high correlation between a decline in temperature and the increase in ozone. The increase in the greenhouse gases in the troposphere raises surface temperatures, while it also reduces temperatures in the stratosphere. The Antarctic ozone hole in lower stratosphere becomes more active; however it will play a role in the reduction of ozone near 40 km altitude. Temperatures at the upper stratosphere at mid-latitudes remain constant, and at lower temperature after the 1990s [WMO, 2007; Steinbrecht et al., 2009]. In the Antarctic region, it appears that a decline of temperature still continues with the increased inter-annual variability observed in the past ten years. Continuous study of chemical reaction coefficients, temperature dependence and transport, etc. is needed. Our research is based on Umkehr observations from the Dobson ozone spectrophotometers of high precision that is

required for detection of significant ozone change (recovery) in relation to the decrease in the ODS concentrations.

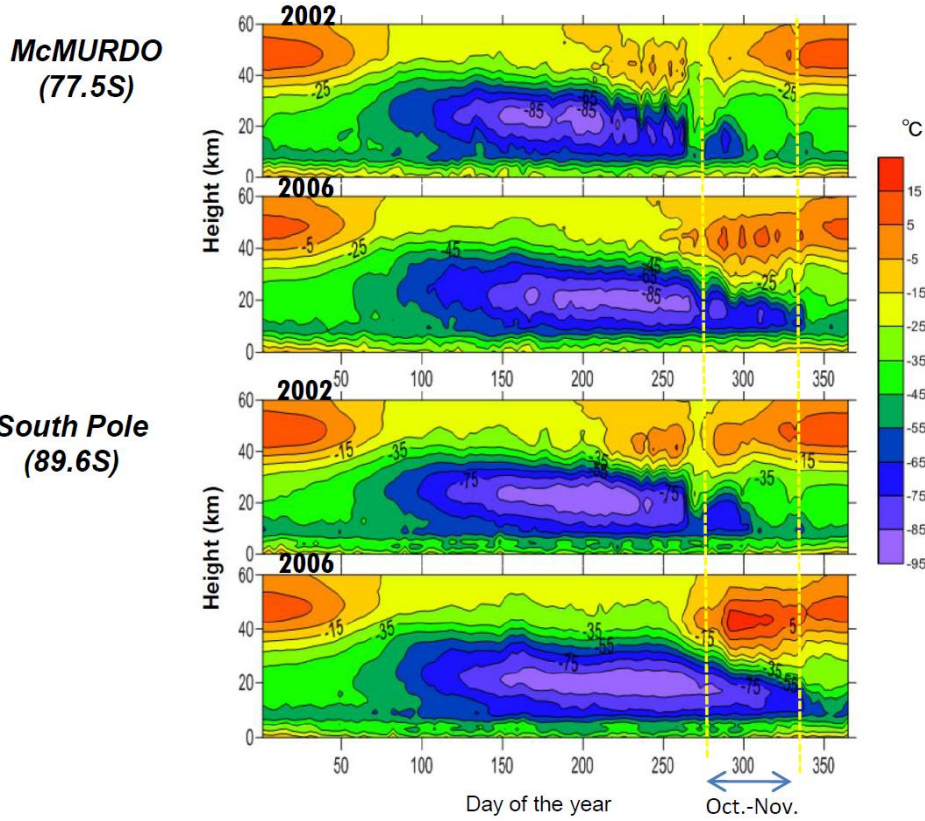


Figure 1 Seasonal change of the stratosphere temperature of two stations in the Antarctic region.

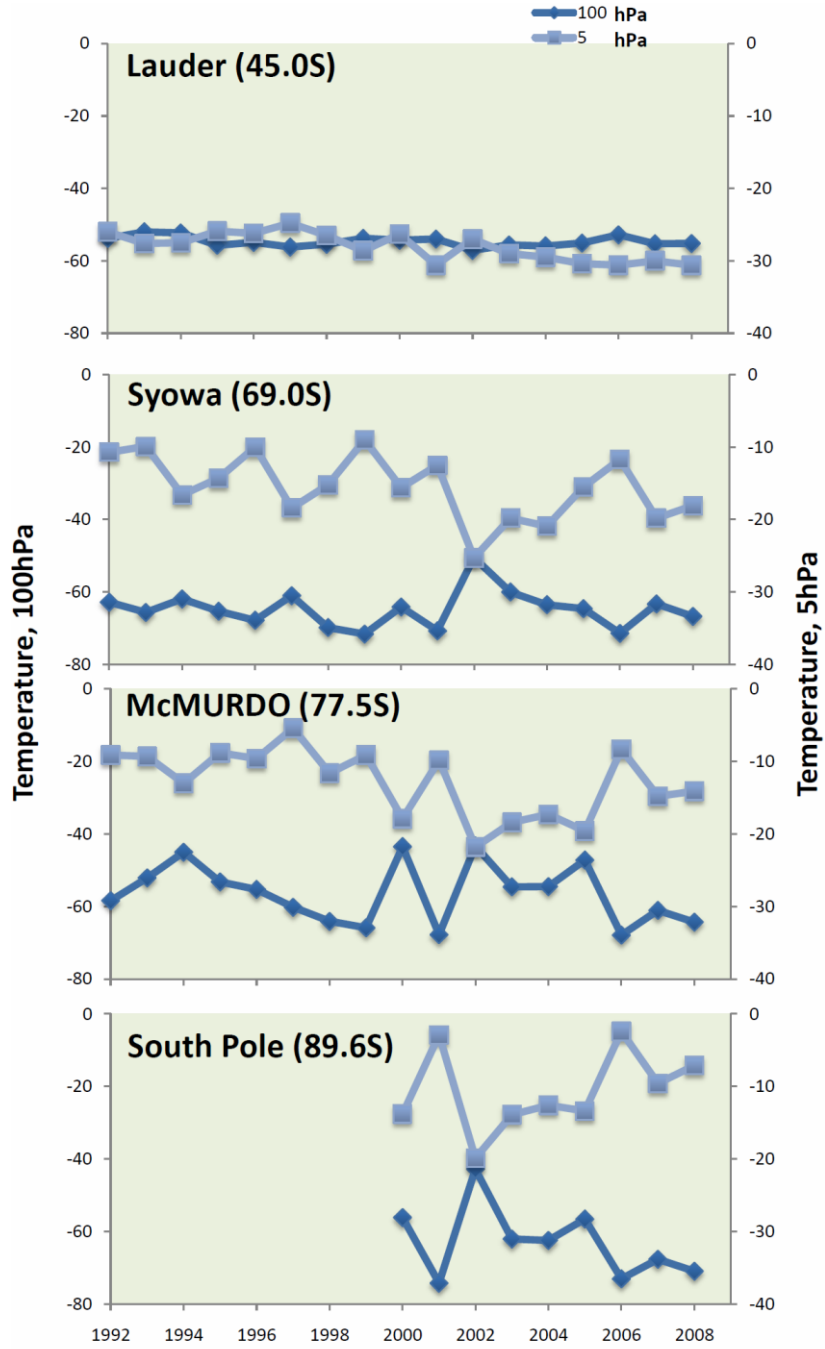


Figure 2 Annual change of the average temperature in November (100 hPa and 5 hPa) at each stations

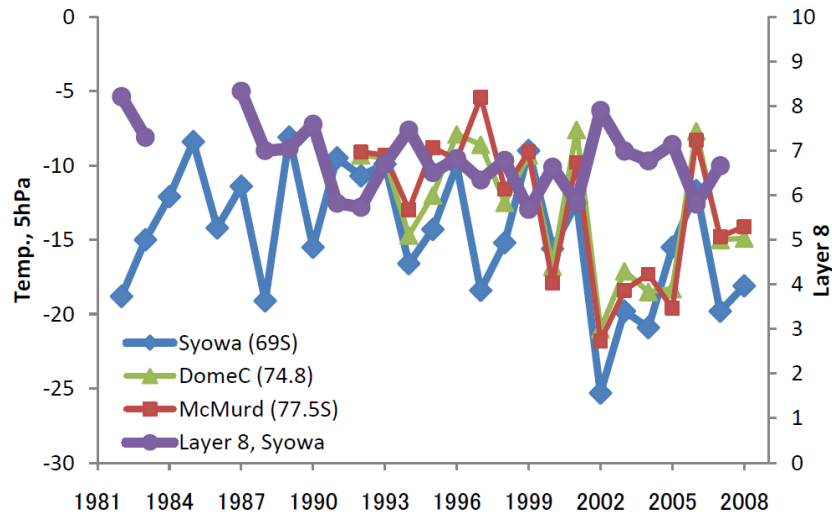


Figure 3 Annual change in November of layer 8 ozone (Syowa) and 5-hPa temperature (Syowa, Dome C and Mc Murdo)

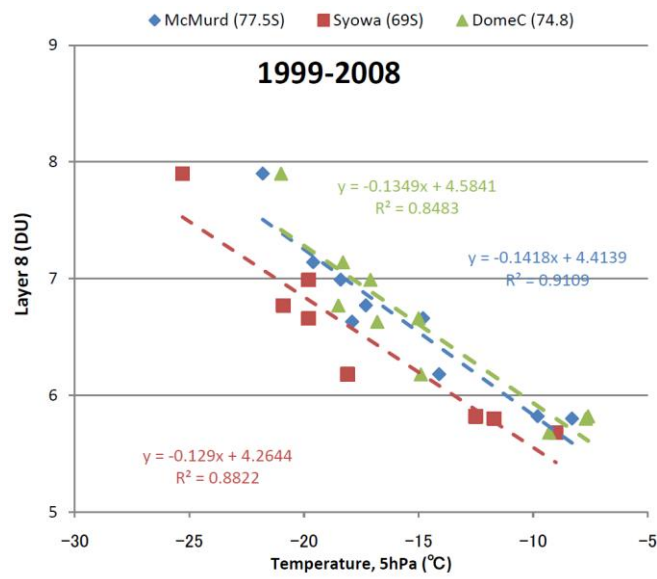


Figure 4 Linear trends of ozone and temperature as well as Figure 3.

The reevaluation of the optical characteristics of individual instruments and correction of Umkehr observations helped to reduce effects of the shifts in the long-term Japanese network data. The re-evaluated Umkehr ozone profiles were verified against independent observational data. Moreover, the use of an a-priori profile independent of the total ozone variability in the Umkehr retrievals (UMK04 algorithm) has optimized ozone dataset for analysis of the long-term trend in stratospheric ozone.

In Chapter 1, the Umkehr measurement data were discussed by introducing new techniques to remove instrumental uncertainty in measured N values by assessing each instrument's characteristics on the basis of intercomparisons with a reference Dobson instrument. Reevaluated and original N values were processed by the Umkehr retrieval algorithm UMK04 as well as by UMK92 to derive vertical ozone profiles. It was found that the ozone profile retrieved by UMK04 shows a clear decreasing trend in the 1980s in the lower stratosphere. Vertical ozone profiles retrieved from revised N-value data sets were compared with coincident ozonesonde and lidar measurements on a daily basis. The results show that the Umkehr deviation from ozonesonde profile is less than 5% in layers 4 to 6, which suggests that reevaluated Umkehr data provide high-quality ozone profiles available for long-term trend analyses. However, more than 10% difference between sonde and Umkehr ozone is found in the lowermost layer. This is considered to be due to KC ozonesondes characteristics (though limited information in this Umkehr layer may be part of the disagreement). Recent colocated data comparisons with the tropospheric lidar system in Tsukuba, display improved agreement with the Umkehr results. Long-term ozone trends in Umkehr measurements show the largest decreases in the upper stratosphere (layers 7 and 8 for Sapporo and Tsukuba). Decreases in the lower stratosphere (layer 4) between 1979 and 1996 are also detected at Tsukuba and Sapporo stations. A decrease in total ozone in this period reflects a decrease in the layer 4 ozone. Results of this study show that the new intercomparison procedure of Umkehr measurements with the fixed reference standard (such as 083 and 116), as introduced in this paper, can provide the international Dobson network with a great improvement of data quality.

In Chapter 2, the long-term ozone trends in re-evaluated Umkehr datasets were discussed in detail. The ozone hole in 2006 was the largest depletion ever observed due to the remaining high concentration of ozone depleting substances in the stratosphere and the lower than usual stratospheric temperatures in the Antarctic region related to the weaker than usual planetary wave activity. In this analysis, components of natural ozone variations related to solar activity and QBO are removed. Ozone in spring season shows clear and steady decreasing both in the upper (9.6%/decade) and the lower (16.6%/decade) stratosphere. On the contrary, ozone in the summer shows only small decreasing in the upper stratosphere and no clear decreasing in the lower stratosphere. Umkehr data are expected to provide important information about stratospheric ozone trends in the Antarctic region in addition to the satellite data. The independent data source from Umkehr measurements is also expected to validate satellite observations. Re-evaluated new Umkehr datasets contributes to the ozone layer assessment in regards to the Montreal Protocol success.

In Chapter 3, the long-term trend and the seasonal characteristic in upper stratospheric ozone is discussed. In the upper stratosphere over Japan, the long-term ozone depletion is seen at higher latitudes (Sapporo and Tsukuba). At Syowa in the Antarctica, a linear trend flattened in the period 1996-2005 after a decrease (-7.9%/decade) in 1977-1996, though recent data show a decreasing trend again. Very low ozone in the higher latitude upper stratosphere (Sapporo and Syowa) is noticeable in recent years. In the lower stratosphere, clear ozone decreases were found at Sapporo (-6.5%/decade) and Syowa (-31.9%/decade) from 1970s to 1996, however, after 1996 trends had been reduced to close to zero. These features agree with changes in total ozone. The trend analyses in the upper and lower stratosphere in spring and summer at Syowa show that a decreasing trend is generally found except in the lower stratosphere (layer 4) in summer season. Especially a large ozone decrease is observed in the last 3 years (2006-2008) in layer 8+9+10 in spring, which continues in the following summer. The year 2006 was “unusual” as extremely low ozone concentration was observed in most layers (except for layer 1) accompanied by low stratospheric temperatures. The difference in trends for ozone measured outside and the inside of the polar vortex is large in layers 2-5 (the lower stratosphere) and 7-9 (the upper stratosphere). It has been shown [Solomon *et al.*, 1986] that ozone depletion in the upper stratosphere is caused by only gas phase chemistry while in the lower stratosphere it is driven by heterogeneous chemistry. Moreover, a seasonal dependence of the ozone depletion in the upper stratosphere is observed since 1995, where about 10%/decade decrease is detected in spring, and ozone recovery is still very low.

In conclusion, we described the re-evaluation technique used for the Umkehr observations, and updated the ozone trends, especially in the upper stratosphere over the middle latitudes. The continuing long-term ozone destruction in the Antarctic region is described in detail, which provides new information about the ozone loss process in the Antarctic region. The measurement re-analysis technique developed by this study for the analysis of a long-term Umkehr measurements can be used to further study the decline of stratosphere temperature, changes in atmospheric circulation field by climate change, and influence of radiation-forcing for more complete understanding of the complicated ozone variation mechanism in the Antarctic region.