## A-1.3 Statistical Analyses of Variabilities of the Ozone Layer

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**Abstract** To analyze spatial and temporal variation of the ozone layer, this research aims to clarify the characteristics of the seasonal and annual variation of ozone-related species by means of statistical methods using mainly satellite data, ground-based or balloon-borne in-situ data, the meteorological data. We have investigated the effective statistics for analyzing parameters in the ozone layer. Order statistics are effective as indices of a representative value and variation of the parameters. We have newly produced a monthly climatological database in each latitudinal zone by using the UARS sensors' data. By investigating long-term temperature in the lower stratosphere in the wintertime of the northern hemisphere, it was found that the winter in 1996/97 was almost cold, and the one in 1997/98 was relatively warmer. The parameters we have analyzed here clearly posed these temperature tendencies. Among these parameters, the data quality analyses about nitric acid ($HNO_3$) and aerosols related on the satellite sensor Improved Limb Atmospheric Spectrometer (ILAS) have been reported here.

**Key Words** Stratosphere, Seasonal and annual variation, Satellite data, ILAS

1. Introduction

In the ozone layer researches, several kinds of data such as satellite data, aircraft data, balloon-borne measurement data, and ground-based measurement data are used. The data quality and measurement error of these data depends on the sensors. Estimation of the data quality is important to deduce scientific results from these composite measurement data.

National Space Development Agency (NASDA) of Japan launched Advanced Earth Observing Satellite (ADEOS) named "Midori" in 1996. Improved Limb Atmospheric Spectrometer (ILAS) aboard ADEOS had measured high latitudinal ozone layer for eight months from November 1996 to June 1997. In addition, large correlative measurement campaigns for ILAS were held during the ILAS operation period. These validation data have been evaluated the data quality and analyzed. These ILAS and in-situ data are expected to provide useful information about the variation of ozone and the related species. From the research by data quality evaluation and comparing with several parameters, the characteristics of the seasonal and annual variation of ozone-related species will be proved well.

2. Research Objectives

This research aims at the following objectives.

- Data quality estimation by means of statistical methods for each atmospheric parameter
- To investigate relations between dynamical and chemical factors on ozone layer variation
- To analyze seasonal and annual variation of ozone-related species
- To clarify the characteristics (generality and specialty) of parameter variation in each measurement period

3. Research Methods

Using data of Japanese and foreign satellite sensors for ozone layer monitoring, balloon and sonde data, worldwide meteorological site measurement data, reliable information will be extracted through the data quality estimation. We have settled the person in charge of investigating each parameter. He/she dealt with the parameter data for several kinds of the sensors. Each person exchanged information about this research.

4. Results and Discussion

During the three-year research period, mainly the following researches have been done; (1) study on the effective statistics for data quality estimation, (2) investigations of stratospheric ozone variability and their relation between dynamical and photochemical aspects focused on the long-term variation in the northern stratospheric circulation, (3) examination of the quality of the ILAS nitric acid ($HNO_3$) data from the comparison with the balloon-borne measurements, (4) research on stratospheric aerosol effects on the ozone layer especially on the observation of Polar Stratospheric Cloud (PSC) from ILAS data. Results and discussion are described for each research sub-topic.

4.1 Study on the effective statistics for data quality estimation

The atmospheric parameters such as temperature, aerosol extinction coefficient, and gas concentrations have unique characteristics on the data variation. They are non-negative, and sometimes take an extremely large value. Average (mean) and standard deviation are the popular statistics. As the results of this study, order statistics are suitable for the atmospheric parameter representative indices. Order statistics such as median, 10 percentile, and 90 percentile are not affected by extreme values nor by the distortion of the data distribution. We have newly produced a monthly climatological database[1)] from the data of the Upper Atmosphere Research Satellite (UARS) sensors by calculating these order statistics. This database contains the following parameters. $O_3$ and $H_2O$ were calculated from the Microwave Limb Sounder (MLS) version 3 data. $NO_2$, $N_2O$, $CH_4$, $CFCl_3$, $CF_2Cl_2$ were done from the Cryogenic Limb Array Etalon Spectrometer (CLAES) version 6 data. $HNO_3$, $ClONO_2$, $N_2O_5$ were done from the CLAES version 8 data. It is expected that this database is useful for several kinds of atmospheric researches.

4.2 Stability of the polar vortex in the lower stratosphere of the northern hemisphere

For the period of 1990s it has been reported that the ozone depletion, which had been believed to happen only in the southern hemisphere, occurred even in the northern hemisphere. This report describes stratospheric circulation in the polar northern hemisphere and its long-term variability. Attention is paid to the atmospheric condition for the 1997 winter when anomalously low temperature and low ozone were observed. In addition, the cause of depletion is elucidated from the photochemical viewpoint. After 1997, we have had relatively warmer winters. A case study is presented for especially warm March in 1999 when the final warming occurred very early.

The main purpose of this study is to investigate characteristics of stratospheric ozone variability and their relation between dynamical and photochemical aspects using several statistical methods. Among all this study focuses on ozone depletion in the northern hemisphere about which several observational results have been reported recently. Moreover, the long-term variation in the northern stratospheric circulation is investigated.

The main source used in this study is the U. K. Meteorological Office (UKMO) analysis data. The UKMO analyses are global analyses of the troposphere and stratosphere since October 1991, produced by the technique of data assimilation. The pressure levels are decided by p= 1000×10**(-i/6) (i=0, 1, 2, ..., 21). Supplementary stratospheric analysis data compiled at Free University of Berlin are used for long-term coverage.

To investigate minor constituents such as $O_3$, ClO, $HNO_3$ we use data from the Microwave Limb Sounder (MLS) aboard the Upper Atmosphere Research Satellite (UARS). The pressure levels are similar to the UKMO data except that the lowest level is set 100 hPa. Though the MLS data are provided since October 1991, data availability is shortened owing to the degradation of power supply. However, some averaging enables us to see longer time-scale variations such as inter-annual variability.

### 4.2.1 Long-term variation in the northern stratosphere

Recently it has been pointed out that the lower stratospheric temperature is decreasing in association with the ozone decrease during the winter and spring time in the polar stratosphere (cf. WMO, Scientific Assessment of Ozone Depletion: 1998[2)]). Investigating the Berlin data over 30 years, we have found the following characteristics.

(1) Temperatures during summer and early winter (July to November) are steadily decreasing; for example, it is about 1K decrease for 10 years.

(2) In January and February, year-to-year variability in temperature is very large in relation to the sudden warming, and the cooling tendency is not clear. However, it seems that the occurrence of major warmings gets less.

(3) In March the cooling tendency is clear, but in May it shows warming tendency in 1990s. This suggests that the seasonal evolution into summer is delayed and that the intensity of final warmings gets stronger.

With regard to the cooling trend in summer (1), it might be due to the global cooling in the stratosphere in conjunction with the global warming in the troposphere; however, WMO (1998)[2)] pointed out an additional effect from the ozone decrease even in the summer time.

On the other hand for the winter and spring cooling (2), (3), it is true up to 1997, but in 1998, 1999 we have had relatively warmer winters (see Figure 1). Especially in 1999, the final warming occurred very early (in early March); we will see this case later.

### 4.2.2 Stratospheric condition in 1996/97 winter

The northern winter in 1996/97 has been reported to be characterized as a stable and prolonged polar vortex and resulting ozone depletion. Zonal mean temperature in the lower stratosphere for the northern hemisphere shows basically similar seasonal march to that in the southern hemisphere. As well known, however, the time of transition into summer circulation occurs earlier in the northern hemisphere than in the southern hemisphere even in this cold winter. During the 1996/97 winter there happened no stratospheric major warming.

The stable and prolonged polar vortex can easily seen on maps for the temperature, wind and potential vorticity fields. During February and March, we can see such robustness of the vortex as to the southern hemisphere with a cold core surrounded by a warm pool having a horseshoe shape. Corresponding to this strong vortex, the edge of the polar vortex is sharply defined.

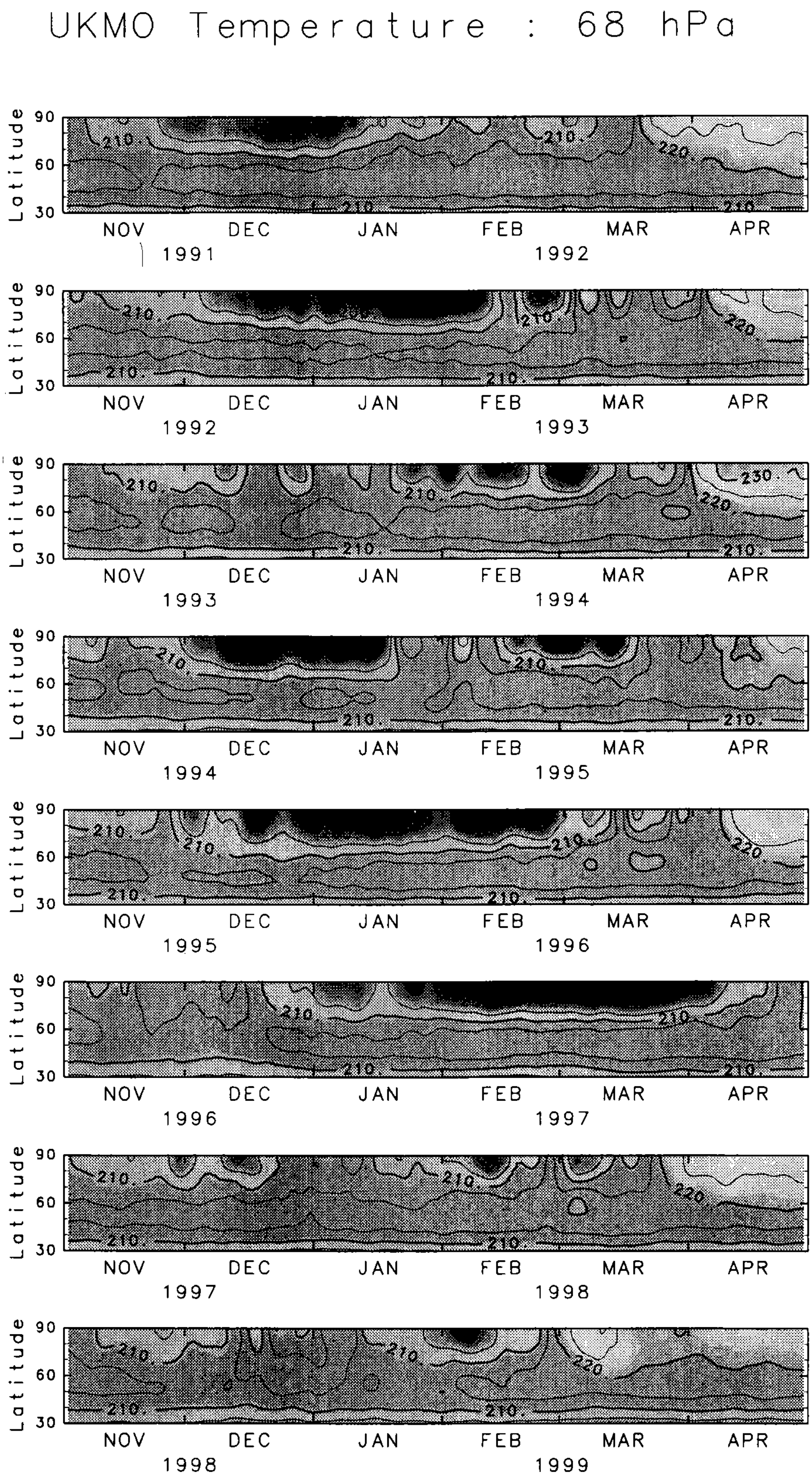


Fig. 1. Temporal-latitudinal cross-section of UKMO zonal mean temperature at 68 hPa. From 1991/1992 wintertime to 1998/1999.

### 4.2.3 Distribution of minor constituents

Using UARS/MLS ozone data, ozone hole like phenomena and corresponding decrease in $HNO_3$ can be seen in 1997. The observation of ClO by MLS is one of the most important components to clarify ozone destruction process in the lower stratosphere. Looking at year-to-year variations of the average ClO distribution in February and March, it is clear that the strong enhancement of ClO was observed in 1997 resulting in the anomalous low ozone in March 1997. These evidences confirm that the depletion of ozone in 1997 spring is definitely due to the photochemical processes.

### 4.2.4 Stratospheric condition in 1998/99 winter

To 1997, the cooling trend seems to be dominated in March, but in 1998 and 1999 we have had relatively warm condition. Especially in 1999, the final warming occurred in early March. In a series of maps for the temperature, wind and potential vorticity fields we can see clear polar vortex in mid February, but it breaks up in early March 1999.

### 4.2.5 Discussion

It has been shown that the ozone depletion in the northern hemisphere in recent years is due to the photochemical ozone destruction. In relation to this ozone depletion, it is also observed that temperatures in the polar lower stratosphere are decreasing. This cooling tendency is a key to understand the ozone depletion, but we still need a missing ring about the polar stratospheric clouds between the ozone destruction and the cooling tendency.

The cooling tendency ceased in 1998 and 1999. The year 1998 is just after the large El-Nino event and we may think of its relation to changes in the tropospheric circulation and relating oceanic variability.

## 4.3 Examination of the quality of the ILAS nitric acid ($HNO_3$) data

The ILAS is a solar occultation satellite sensor which measures the absorption of stratospheric species in the infrared region of solar radiation[3)]. It was launched on board the ADEOS satellite in August 1996. Regular measurements were made between November 1996 and June 1997. Vertical profiles of nitric acid ($HNO_3$) were retrieved using all of the $HNO_3$ absorption lines appeared in 850 – 1610 $cm^{-1}$ region which ILAS measured. Among these lines, a contribution from the $\nu_5$ and $2\nu_9$ bands at 879 and 895 $cm^{-1}$ was largest. The contributions from the $\nu_3$ and $\nu_4$ bands at 1326 and 1304 $cm^{-1}$ were also larger as compared to those from the other lines. $HNO_3$ values were retrieved every 1 km, although the actual vertical resolution was 2 km.

ADEOS/ILAS validation balloon campaigns were carried out from Kiruna, Sweden (68°N, 21°E) in February and March 1997 and Fairbanks, Alaska (65°N, 148°W) in April and May 1997[4)]. In this study, the quality of the ILAS $HNO_3$ data was examined from the comparison with the balloon-borne measurements.

### 4.3.1 Balloon-borne measurements

From Kiruna, four balloon-borne instruments measuring $HNO_3$ or $NO_y$ were flown a total of six times. From Fairbanks, three instruments were flown, once each. Except for the Chemiluminescence Detector (CLD) in-situ total reactive nitrogen ($NO_y$) measurements, all of the measurements were performed by remote sensing using absorption or emission spectra of $HNO_3$ in the infrared region.

The CLD instrument measured $NO_y$ (= $NO + NO_2 + NO_3 + HNO_3 + 2(N_2O_5) + HO_2NO_2 + ClONO_2 + BrONO_2$ + aerosol nitrate) using the $NO/O_3$ chemiluminescence technique after the catalytic conversion of the component species into NO on the surface of heated (300°C) gold tubes[5,6]. The amount of $HNO_3$ was calculated from the $NO_y$ value using a box model developed by Atmospheric and Environment Research Inc. (AER) based on the 10-d back trajectories of air masses.

The Cold Atmospheric Emission Spectral Radiometer (CAESR) measures thermal emission from the atmosphere. The Limb Profile Monitor of the Atmosphere (LPMA) is an infrared Fourier transform spectrometer which measures various species from solar occultation spectra. The Michelson Interferometer for Passive Atmospheric Souding-Balloon-borne version 2 (MIPAS-B2) is a cryogenic Fourier transform infrared spectrometer, which measures atmospheric thermal emissions from the limb direction. The far-infrared spectrometer (FIRS-2) is a high-resolution, thermal emission Fourier-transform spectrometer. Two independent profiles were obtained during the 970430 flight by looking in two different directions at similar latitudes separated by roughly 4° in longitude. The MkIV interferometer was designed and built at the Jet Propulsion Laboratory (JPL) in the early 1980's. Its wide spectral bandpass (650-5650 $cm^{-1}$) and high resolution (0.01 $cm^{-1}$) allow the simultaneous retrieval of over 30 different atmospheric gases including all the important members of the nitrogen family. The $HNO_3$ profile obtained on 970508 was retrieved from solar absorption spectra measured from 38 km altitude during sunrise.

In addition to the series of balloon-borne measurements, in-situ measurement of $NO_y$ at 12 km was also made by the Deutsche Zentrum fur Luft- und Raumfahrt (DLR) from the Falcon 20 research aircraft on 970128 and 970130. The measurement technique was same as that for the CLD instrument. The $HNO_3$ values were estimated by assuming that 90 % of $NO_y$ was in the form of $HNO_3$ based on the AER model results for the CLD experiments.

### 4.3.2 Method

In this study, two different ILAS values were compared with the balloon data. First, the nearest ILAS $HNO_3$ vertical profile, which was obtained on the same UT-day, was compared. If the neighboring two ILAS data were obtained within roughly the same distance from the location of the balloon measurement, the data with a potential vorticity (PV) value similar to that of the balloon data was used. The ILAS $HNO_3$ values were interpolated to yield values at the potential temperatures where the balloon data had been obtained. This data will hereafter be referred as the nearest data. The United Kingdom Meteorological Office (UKMO) data were used for the PV calculation. Second, the ILAS values obtained within $\pm 1$ UT-day were selected. Then the ILAS $HNO_3$ values and PV values for each ILAS data point were interpolated to yield values at the potential temperatures where the balloon data had been obtained. When the PV value of the ILAS data point agreed within 10 % with the PV value of the balloon data, these data were used to yield the averaged ILAS profile. This data will be referred to as the PV-averaged data. Since the amount of balloon data, which could be used for the comparison study, was limited, the first approach, using only the nearest data, may have a statistical limitation when the agreement is evaluated. For the second approach, using the PV-averages, 10 to 20 values were generally averaged at each altitude, providing a statistically more robust estimate to confirm the degree of agreement.

### 4.3.3 Results and discussions

The agreement with the nearest values with the balloon-derived values was generally similar to or better than that for the PV-averaged values, suggesting that the results from the nearest data comparison are statistically reliable. The systematic and random differences between the ILAS and balloon measurements were considered to provide an estimate of the absolute accuracy and precision of the ILAS $HNO_3$ measurement. Considering the errors in the balloon-borne measurements and the real difference in the $HNO_3$ profiles at the locations where the ILAS and balloon-borne measurements were made, the absolute accuracy and precision of the ILAS measurements might be better than those estimated from the systematic and random differences.

The random difference between the ILAS and balloon-borne measurements was generally similar at all altitudes. From these random differences, the precision of the ILAS measurement was estimated to be between 0.7 and 0.9 ppbv at altitudes between 15 and 30 km. These values corresponded to 33, 9, and 14 % at altitudes of 15, 20-25, and 30 km, respectively. The precision of the ILAS measurements estimated from the spectral fitting was 24, 15-20, and higher than 50 % at these altitudes. The precision of the ILAS measurement estimated from this comparison study was, therefore, generally better than the instrumental error.

The systematic difference between the ILAS and balloon-borne measurements is evident. The ILAS values between 20 and 30 km were systematically lower than the balloon values and the difference tended to be larger at higher altitudes in accordance with the results of the individual comparisons presented above. The absolute accuracy was estimated from these systematic differences to be as good as 0.5 ppbv (5 %) at 20 km, while a negative bias of 1 ppbv (14-18 %) was indicated at 25-30 km. The absolute accuracy of the ILAS measurements estimated only from the uncertainties in the line parameters was 10 %. Consequently, the disagreement found here was slightly beyond the expected range. As described above, the FIRS-2 and MkIV values decreased by 8 and 4 %, respectively, from those used in this study, when the $HNO_3$ values were retrieved from the same $HNO_3$ absorption lines used by the ILAS. This could be the cause of the systematic difference between the measurements; however, it can not explain the vertical profile of the systematic difference. In addition, although the same $HNO_3$ lines which mainly used by the ILAS were used for the MIPAS-B2 and CAESR retrievals, similar systematic differences were generally seen.

## 4.4 Stratospheric aerosol effects on the ozone layer

Aerosol extinction coefficient data in the stratosphere measured by Stratospheric Aerosol and Gas Experiment II (SAGE II) and ILAS have been analyzed. Especially the sighting probabilities on the observation of Polar Stratospheric Cloud (PSC) from ILAS data were calculated and analyzed. Summary of the results is described as follows.

The four-wavelength extinction coefficients of stratospheric aerosols observed with SAGE II were analyzed. SAGE II is an occultation sensor which have measured aerosol extinction at 1.02, 0.525, 0.453, 0.385 μm from 10 km up to 45 km in altitude since 1985. The time series of 11-year record from 1985 through 1995 shows the strong increase of the extinction in 1991 after the eruption Pinatubo. The wavelength dependence of the extinction coefficient also significantly changed after the eruption of Mt. Pinatubo (June 1991, Philippine), showing the existence of large particles. Even in the background period before the eruption, the wavelength-dependence show significant spatial variation. The wavelength dependence would be good information to investigate the micro physical evolution processes of aerosol particles with the air mass transportation in the lower stratosphere.

The ILAS on board the ADEOS satellite observed many Polar Stratospheric Cloud (PSC) events during the winter of 1997, when there was a significant decrease in Arctic ozone. More than a hundred PSC profiles were identified over the Arctic during the period from November 1996 to March 1997. In January and February most PSCs appeared around the Greenwich meridian at an altitude of about 23 km, while in March they appeared much further east, at approximately 120°E, and at lower altitudes. PSC sighting probabilities were calculated as the ratio of the number of PSC events to the total number of measurements for every altitude and every 10-day period. The highest value of the sighting probability was 0.5 in mid-January. Movement of the PSC locations was strongly connected with the displacement of the polar vortex and cold air mass. The PSCs in the 1997 Arctic winter were characterized by their prolonged appearance until mid-March, associated with the long-persisting polar vortex. Most of the PSC events took place under low-temperature conditions below the NAT (Nitric Acid Trihydrate) temperature, but some PSCs were seen at above this temperature. Although there are uncertainties regarding the accuracy of the UKMO temperature and the NAT temperature estimation, the possibility of warm PSCs was presented.

Aerosol extinction at 780 nm (version 3.10) from ILAS was compared to measurements by three balloon-borne aerosol sensors flown during the winter of 1997 at Esrange, Kiruna, Sweden (67.9°N, 21.1°E). The six comparisons of ILAS extinction with extinction calculated from in-situ optical aerosol counters indicated relative differences of up to 100%. The agreement with one remote sensing instrument showed a much better agreement to within 20% below 18 km. The results of this validation exercise confirm the quality of the ILAS aerosol data and their potential for scientific use; however, there are still some uncertainties in the ILAS data that should be addressed in future versions.

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