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学位論文題目	A study on depletion of the upper stratospheric ozone in the Antarctic from Umkehr ozone profile	
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論文内容の要旨

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 Shimatzu 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.

博士論文の審査結果の要旨

論文題名（和訳）は「反転観測による南極域上部成層圏オゾン破壊に関する研究」というもので、本論文は既刊の３編の学術誌掲載論文をまとめたものである。第１章は、反転オゾン観測に含まれる測定誤差の問題を扱い、誤差の補正方法を提案、他の観測手法であるオゾンゾンデ等と比較して測定精度の検証を行った。第２章では、1977 年から 2007 年まで南極昭和基地で観測された約 1300 の反転オゾンプロファイルから、その長期トレンドと季節の特徴を調べた。第３章では、南極昭和基地を含む、札幌、つくば、那覇のわが国の反転オゾン観測網について、測定手法の再評価と新しいアルゴリズムによる再解析を行い、長期の成層圏オゾンの変化傾向を論じた。１月７日午後、論文公開発表会に併せ本審査委員会をもち、下記の通り、博士学位論文として認めることとした。

南極上空のオゾンホールについては、2006 年に過去最大規模の成層圏オゾンの減少が観測される等、2000 年代に入っても依然として深刻な状況にあることが指摘されている。これまでの地上観測および人工衛星観測から、南極上空でのオゾン全量は、1980 年代から 1990 年代前半にかけて大きく減少したことが知られているが、そのオゾン層破壊の主な要因はフロン等のオゾン層破壊物質の存在であり、下部成層圏の極渦内の低温条件下での物理・化学過程、即ち極成層圏雲粒子上での不均一反応が関与している。一方、高度 40 km 付近の上部成層圏でのオゾン破壊は、フロン等の分解によってもたらされた塩素原子が触媒となってオゾンを破壊する気相反応によるもので、1974 年、ローランドとモリナによって初めて指摘されたものである。上部成層圏のオゾン濃度の観測は、様々な人工衛星によって 1979 年以降全球的に行われているが、極域には及んでいない。その結果によるオゾンのトレンドは、特に高緯度側で減少率が大きく、北半球より南半球で大きな減少を示している。人工衛星による観測が難しい南極域では、極渦強度、その持続期間などがオゾンの年々変動に密接に関連しているが、極渦内部での上部成層圏オゾン減少の実態を把握するためには、長期的な変動ならびに減少率を観測データから定量的に見積もることが必要である。反転観測は成層圏オゾン全体の鉛直分布を理解する上で貴重な情報を提供するものである。

本論文は、ドブソン分光光度計を使い、紫外線の天頂散乱光の二つの波長での強度比の太陽高度角依存からオゾン鉛直分布をインバージョン法によって求める「反転観測法」をより精緻化し、南極上空の上部成層圏オゾンの変動傾向を初めて導出したものである。反転観測法は、かなり高度な技術を要するリモートセンシング手法であり、現在では世界中で 20 カ所でしか観測がなく、南極域では昭和基地が唯一の観測点となっている。申請者は、近年新しく開発された反転観測アルゴリズム（UMK04）を利用し、実際の観測結果を解析する上での様々な補正処理手法を考案し、年々の長期変化傾向を議論できる高精度のデータ導出手法を確立した。主な誤差要因になっている太陽天頂角依存性について、測器毎の特性を調べデータ処理の評価・改善を行い、また、アプリオリなオゾン鉛直分布を、観測日、緯度、気圧を関数として最適化された近似式により決定するなどの方法である。この手法を昭和基地での 1977 年以来のデータに適用し、上部成層圏の高度 40 km 層におけるオゾン量の変化傾向を明らかにした。その結果、この 30 年、一貫して減少傾向にあることが示された。これは、従来中緯度での結果から言われてきた、1996 年以降は

減少傾向が止まり、回復傾向が見られ、フロン規制により、オゾン破壊がなくなってきたという説明が南極域ではなりたっていないという結果を初めて導いた。

以上のように、本論文は、反転オゾン観測データから、新しい手法を駆使して、これまでよりも正確なオゾン濃度の導出を行い、それらのデータを基に南極域のオゾン層破壊についての検討を行った。特に今までよく知られていなかった上部成層圏におけるオゾンの減少傾向のより定量的な理解がなされ、季節変化の特徴および長期的変化傾向について、新たな知見を提供した。このように、極めて重要な論文であるとして、博士学位論文にふさわしいことを全員一致で認めた。