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ILAS observations of chemical ozone loss in the Arctic vortex during early spring 1997

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Abstract. Chemical ozone loss rates were estimated for the Arctic stratospheric vortex by using ozone profile data (Version 3.10) obtained with the Improved Limb Atmospheric Spectrometer (ILAS) for the spring of 1997. The analysis method is similar to the Match technique, in which an air parcel that the ILAS sounded twice at different locations and at different times was searched from the ILAS data set, and an ozone change rate was calculated from the two profiles. A statistical analysis indicates that the maximum ozone loss rate was found on the 450 K potential temperature surface in February, amounting to 84 ppbv/day. The integrated ozone loss for two months from February to March 1997 showed its maximum of 1.5±0.1 ppmv at the surface that followed the diabatic descent of the air parcels and reached the 425 K level on March 31. This is about 50% of the initial (February 1) ozone concentration. The present study demonstrated that data from a solar occultation sensor with a moderate altitude resolution can be used for the Match analysis.

Introduction

Quantitative analyses of the amounts of ozone destruction and/or ozone loss rates in the polar vortex have been done using TOMS data, satellite-borne ozone profiling sensor data such as MLS, and ozone sonde data at particular ground stations [*Newman et al.*, 1997; *Manney et al.*, 1997; *Krcher et al.*, 1999]. These analyses, however, have not been able to clearly discriminate chemical ozone destruction due to reactions involving chlorine and bromine that originate in anthropogenic compounds such as chlorofluorocarbons from dynamical ozone changes caused by advection and mixing through the vortex boundary. To solve this difficulty, *von der Gathen et al.* [1995] and *Rex et al.* [1997, 1998, 1999] applied a new technique called "Match" to a data set of ozone sonde measurements. They calculated the amount of ozone change in an air parcel that has passed two separate locations inside the polar vortex so that dynamic effects can be neglected and only chemical changes remain.

von der Gathen et al. [1995] and *Rex et al.* [1998] estimated ozone loss rates from ozone sonde data obtained in the Arctic region during the winter of 1991/1992. They analyzed cases in which air parcels were observed accidentally by two (or more than two) ozone sondes. On the other hand, in the "Match" experiment of *Rex et al.* [1997, 1999], the flight path of an air parcel that has passed an ozone sonde measurement at a certain location was predicted with a trajectory analysis technique, and another ozone sonde was released at a location on the flight path or close to it in order to measure the same air parcel. *Rex et al.* [1999] and *Rex et al.* [1997] made a number of ozone sonde measurements based on trajectory analyses in the Arctic winter of 1994/95 and that of 1995/96, and derived temporal changes of ozone loss rates as a function of altitude (potential temperature). By integrating the ozone loss rates, they calculated total amounts of ozone loss in each winter/spring. Since this technique avoids dynamic effects, the derived quantities are regarded as the amounts of ozone destroyed purely chemically.

It is quite natural to apply the similar technique to ozone profile data obtained with satellite-borne sensors. The Improved Limb Atmospheric Spectrometer (ILAS) developed by the Environment Agency of Japan (EA), and on board the Advanced Earth Observing Satellite (ADEOS) which was launched by the National Space Development Agency of Japan (NASDA), made intensive measurements over high-latitude stratospheres, totaling 731 sunrise events over a relatively narrow latitude range of 67 to 70 degrees N during the period from February to March in the Arctic spring of 1997, when a relatively distinct polar vortex was persisting. Therefore, a number of air parcels are likely to have been unintentionally sounded at different ILAS measurement locations. Although we must rely on accidental coincidences in the same way as *von der Gathen et al.* [1995] and *Rex et al.* [1998], there is a sufficient number of coincidences to apply a similar analysis to that of *Rex et al.* [1997, 1999] as will be shown.

This paper presents preliminary estimates of the ozone loss rates and amounts based on a technique similar to that of *von der Gathen et al.* [1995] and *Rex et al.* [1998] using the ILAS ozone data for the spring of 1997 and the trajectory analyses with the UKMO assimilation data. It is noteworthy that satellite sensor data with a relatively low vertical resolution (ILAS has a vertical resolution of about 2 km) can provide reasonable results with the present analysis method.

Data and analysis method

ILAS is a sensor that is based on the solar occultation technique and provides vertical profiles of gas species by infrared spectrometry [*Suzuki et al.*, 1995]. ILAS was on board ADEOS and obtained data for a total of 6,743 sunrise (in the northern hemisphere) and sunset (in the southern hemisphere) solar occultation events during a period of 8 months from November 1996 to June 1997 [*Sasano et al.*, 1999a]. Measurement targets were profiles of ozone, nitric acid, nitrogen dioxide, methane, nitrous oxide, water vapor, and aerosol extinction coefficient at 780 nm from about 10 km up to 60 km depending on parameters. The present analysis used the ozone data from Version 3.10 products, which has been validated against ozone sonde and HALOE ozone measurements [*Sasano et al.*, 1999b; *Lee et al.*, 1999].

The following procedure is used to search for pairs of ILAS measurements that are considered to have sounded the same air parcel. As the ILAS ozone data is provided as a function of geometrical altitude, the vertical coordinate is transformed to a potential temperature coordinate by using UKMO assimilation data [*Swinbank and O'Neill*, 1994]. Ozone mixing ratios on potential temperature surfaces are calculated by a cubic spline interpolation with a 25 K inter-

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val (corresponding to about 1 km in altitude interval) from 375 K to 550 K. The flight path of an air parcel observed initially at a certain location (Location #1) for each ILAS measurement is calculated for a period of 10 days with a trajectory calculation program (supplied by R. Swinbank) and the UKMO data. The trajectory analyses are carried out along an isentropic surface for each air parcel having a potential temperature from 375 K to 550 K with 25 K intervals. The UKMO data is linearly interpolated with distance horizontally, with time temporally, and with potential temperature vertically. The integration in calculating trajectory is done every one hour with the third-order Runge-Kutta method. Results of trajectory calculations are stored as latitude, longitude, temperature, and pressure of the air parcel every hour.

When another ILAS measurement is made on the trajectory of the air parcel or close to it (the definition of closeness is discussed later) at the time just when the air parcel passed, the air parcel is considered to be sounded again by the second ILAS measurement (Location #2). Thus, a pair of measurements that gives the same air parcel sounding by ILAS is defined. The ILAS time is rounded up to every hour sharp, considering the fact that the UKMO data is provided once every day at 12 UT.

Descent due to diabatic cooling is taken into consideration by changing the potential temperature of the moving air parcel [*Rex et al.*, 1998; 1999]. The cooling rates averaged over the polar vortex were taken from Figure 1 of *Knudsen et al.* [1998]. The ozone mixing ratio of the air parcel sounded at Location #2 is modified and given by a value at the level of changed potential temperature. Note that wind velocity and direction data on the pre-assigned potential temperature surface are used for trajectory calculations, neglecting their changes due to diabatic descents.

Trajectory analyses are carried out for all the ILAS measurement events to identify pairs of measurements of the same air parcels. There are cases when one air parcel is observed not only twice but in multiple events. In the present analysis, those cases are also treated as independent pairs.

Pairs of measurements are searched for from 731 events in total observed during the period from February 1 to March 31 in the Northern Hemisphere. Since chemical destruction of ozone is considered to occur mainly inside a polar vortex, the search was done only for the pairs obtained inside the polar vortex. In order to discriminate the data, the poleward edge of the vortex boundary was defined on each day [*Kreher et al.*, 1999; *Bodeker, G., private communications*, 1998] based on the definition by *Nash et al.* [1996] for a daily potential vorticity map on each potential temperature surface. In this way the ILAS measurements that were located well inside the polar vortex were selected for further analysis as described below.

The total time that the air parcel spent from the start to the arrival is calculated. The time that the air parcel was lit by the sun (sunlit time) is also calculated from solar zenith angle information [*Knudsen et al.*, 1996]. The total time and the sunlit time are stored as attached information for each air parcel as well as the minimum temperature, and the maximum and minimum potential vorticity that the air parcel experienced.

The criterion for deciding whether an air parcel starting at a certain location comes close to another location, in other words, whether the air parcels observed at two different locations can be considered to be identical, was chosen on the basis of the analysis similar to that by *Rex et al.* [1998, 1999]. Figure 1 shows the standard deviation of ozone change as a function of criterion distance, where the ozone change is corrected by the expected ozone change along the trajectory. The expected ozone change was calculated by multiplying the sunlit time along the trajectory with the ozone change rate per sunlit time. The way of determining the ozone change rate is described below. If a larger distance was used, residuals from the regression line became more scattered, thus suggesting that different air parcels were seen by the ILAS. In this way, 400 km was chosen as the criterion for the present analysis, and a total of 7,171 air parcels (pairs of measurements) were identified for the period from February to March between 375 K and 550 K surfaces at a 25 K interval.

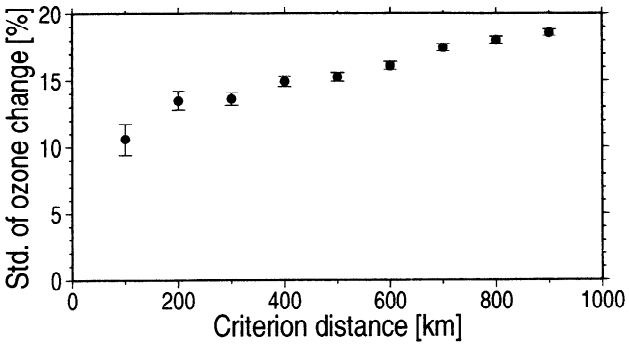

Figure 1. Standard deviation of ozone change as a function of criterion distance. The figure was produced for the data set for the period from March 1 (day 60) to March 31 (day 90) at 475 K potential temperature level.

In addition, changes in potential vorticity along the air parcel trajectory were investigated in the same manner as the above. If a change in potential vorticity, which is originally a conservative quantity, is significantly large, there must have been a problem in selecting the pairs due to mixing of independent air parcels with different potential vorticities, large uncertainties in the trajectory calculation, and so on. To smooth out rapid fluctuations that may be due to potential vorticity calculation errors, a 24-hour running mean was first applied to the potential vorticity data. In the present analysis, the criterion for potential vorticity change, as defined as the difference between the maximum and the minimum along the same trajectory divided by their mean, was set as 15% along the air parcel flight path, and those pairs having potential vorticity changes larger than that were discarded. Finally, a total of 2,816 pairs were selected for further analysis. No distinct trends were found in the potential vorticity changes with time along the trajectory, suggesting no systematic effects of transport. For reference, *Rex et al.* [1998] and *Rex et al.* [1999] adopted the criterion of 500 km for distance, and 40% and 25% for potential vorticity change.

By applying the method described above, the dates and times when the ILAS measurements were made at Locations #1 and #2, amount of ozone change, total time, and sunlit time are calculated and stored for each pair of measurements. From these, the ozone change rate can be evaluated both for the total time and the sunlit time. We cannot, however, tell exactly when the ozone loss process happened for each air parcel. A statistical treatment is applied to the subset of measurement pairs to reduce scatter of ozone change rate results in the following way as was done in *von der Gathen et al.* and *Rex et al.*

Firstly, all the pairs are selected under the condition that the starting date is within 7 days before each target day and the arrival date is within 7 days after it. Secondly, assuming that ozone changes are linearly proportional to the sunlit time, a proportional coefficient (ozone change rate against sunlit time) is calculated using the least-squares method. These calculations are made for all potential temperature

surfaces and for all days from February 1 to March 31. The final results are ozone change rates for sunlit time as a function of potential temperature and date.

Using the ozone change rates for sunlit time, ozone change rates per day can be estimated by multiplying them by the sunlit time (hours) for each day. The sunlit time (hours) for each day can be calculated as the average for air parcels of interest from the data on sunlit hours over each trajectory. In addition, the net ozone change for two months can be evaluated by integrating the ozone change rate per day with time, taking into consideration the altitude change due to descending motion of the air parcels.

Results and discussion

Figure 2 (left panel) shows the final results of ozone change rates per day for sunlit time as a function of potential temperature and date. The solid and dotted curves in the figure represent the isopleths of 99% and 95% statistical significance. The appearance of PSCs (polar stratospheric clouds) is also indicated with red circles in the same figure. The 780 nm aerosol extinction coefficient analyses were done by *Hayashida et al.* [Arctic polar stratospheric clouds observed with the Improved Limb Atmospheric Spectrometer during the winter of 1996/1997, submitted to *J. Geophys. Research*, 1999] to detect PSC appearance. The circles in the figure indicate when and where the PSCs appeared at any of the ILAS measurement locations for each day.

The local maximum ozone loss rate is found on the 450 K surface in the latter half of February, amounting to 84±17 ppbv/day. Significant negative regions correspond well to those for PSCs appearance as well as for very low temperature [*Terao*, 1999]. This strongly suggests that the well-recognized ozone destruction mechanism acted on the air parcels, in which generation of PSCs, activation of chlorine species due to heterogeneous reactions on PSC surfaces, and chain reaction that destroyed ozone under a sunlit condition occurs in sequence. Positive ozone changes are seen at some altitudes and for some periods although they have no statistical significance. Detailed results including HNO_3 changes are reported in *Terao* [1999] based on extension of the present analysis to other species.

The right panel of Figure 2 shows the integrated ozone change as a function of potential temperature from day 32 (February 1) to day 90 (March 31) with exceptions of day 41-90 and day 64-90 for the uppermost two levels, respectively. The integrated ozone loss shows its maximum of 1.5±0.1 ppmv on the surface that reached the 425 K level on March 31 from the 457 K level on February 1 due to the diabatic descent of the air parcels. This is about 50% of the initial (February 1) ozone concentration.

An ozone decrease was estimated by *Kreher et al.* [1999] for the period from February 1 to March 25 using ozone sonde data obtained over Kiruna (67.9 N, 21.1 E) as a part of the ILAS validation balloon campaign [*Kanzawa et al.*, 1997]. With diabatic descent rates incorporated in this analysis, a maximum of 0.63 ppmv/month (a total of about 1 ppmv, 33%) ozone loss was derived for the air descending inside the polar vortex from 475 K to 460 K for that period. This analysis used long-term observation data made at a particular single location, thus likely suffering from the combined effects of both transport and chemistry on the derived ozone loss amounts.

On the contrary, an analysis for the same Arctic winter was made by *Knudsen et al.* [1998] using ozone sonde data obtained at many locations and accounting for horizontal transport by assessing the exchange of air parcels across the vortex edge with domain filling trajectory calculations. They reported ozone decreases of 1.45 ppmv, 1.59 ppmv, 1.24 ppmv, and 1.20 ppmv for the period from January 6 to

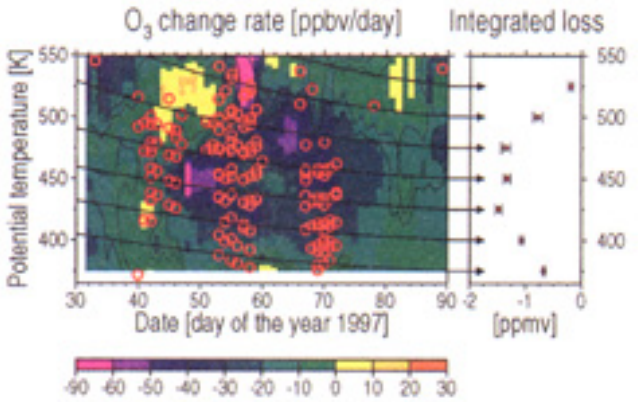

Figure 2. (Left) Color-coded ozone change rates (in ppbv per day) as a function of potential temperature and date (day of the year 1997), which were vertically interpolated into data with 1 K interval by a cubic spline. Solid and dashed curves indicate statistical significance of 99% and 95%, respectively. Red circles indicate the appearance of PSCs (see text). Smooth curves show altitude (potential temperature) changes of air parcels that finally reach the altitude (potential temperature) levels indicated on March 31 [adopted from *Knudsen et al.*, 1998]. (Right) Integrated ozone changes from day 32 (February 1) to day 90 (March 31) (day 41-90 and day 64-90 for the uppermost two levels, respectively) along the smooth curves showing the altitude changes. Error bars represent one sigma.

April 6 for the air masses on 425 K, 450 K, 475 K, and 500 K potential temperature surfaces at the end of the period, respectively. The chemical ozone loss at the 425 K surface estimated as 1.5 ppmv in our case agrees well with the results by *Knudsen et al.* [1998].

Since a solar occultation sensor, in general, has a large measurement volume (approximately 2 km vertical and 300 km in line of sight), it must be carefully examined if the separation of chemical and dynamical effects is complete and if no systematic error is introduced. When an air parcel with such a large volume consists of independent air masses coming from different geographical regions separated far away a few days earlier, the ozone loss derived may be affected by that. Any transport-induced change in ozone should occur independently from sun light. To check this, we conducted a bivariate regression analysis that includes a term allowing for ozone loss in the dark as well as a term in sunlight [*Rex et al.*, 1998, 1999] to see whether any change of ozone is found during darkness. The results show that the ozone loss rates from February 9 to March 1, on the 450 K potential temperature surface are −5.3±4.0 ppbv per sunlit hour and 0.3±2.2 ppbv per dark hour. This indicates that the ozone did not significantly change during darkness, suggesting that no systematic bias was introduced into the ozone loss calculations by transport (dynamical) effects. It should be noted that a bivariate analysis could not be applied to cases when the two variables (sunlit hour and dark hour, in our case) are highly correlated to each other in March. Further analyses may be necessary to fully investigate the possible errors due to dynamical effects.

The present study has derived quantitatively the chemical ozone loss rates as a function of altitude (potential temperature) and date from the analysis of ozone changes in the same air parcels observed by a satellite-borne solar occultation sensor. This analysis is an extension of those by *von der Gathen et al.* [1995] and by *Rex et al.* [1998] who used the data from ozone sonde observations, to satellite sensor data. It was demonstrated that satellite sensor data that, in general, has a vertical resolution of 1-2 km could

be applied to such an analysis. Since a polar orbital solar occultation sensor such as ILAS can intensively measure high-latitude regions, it can provide a very useful data set for an analysis such as presented in this paper. In addition, ILAS provides data on not only ozone but also other chemical species such as nitric acid, nitrous oxide, methane, water vapor, and aerosol extinction coefficients obtained at the same time and at the same location. This means further comprehensive analysis is possible especially on ozone depletion mechanisms involving heterogeneous reactions on the surfaces of polar stratospheric clouds.

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