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# ILAS observations of chemical ozone loss and changes in nitric acid and nitrous oxide concentrations in the Arctic vortex during early spring 1997

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**Abstract.** Reanalysis was made for quantitative chemical ozone loss rates and amounts in the Arctic stratospheric vortex by using ozone profile data (Version 4.20) obtained with the Improved Limb Atmospheric Spectrometer (ILAS) for the spring of 1997. The analysis method is similar to the Match technique. Changes in concentration of nitric acid and nitrous oxide were also investigated with the same analysis method, taking the advantage of the ILAS multiple species measurements.

## Introduction

Separating ozone changes due to chemistry from those due to dynamics is very important for better understanding of the polar stratospheric ozone depletion. However, that is a difficult task and requires careful treatments. One method to solve this difficulty is known as "Match" [von der Gathen *et al.*, 1995; Rex *et al.*, 1998, 1999]. They use pairs of ozonesonde profiles which are obtained at separate locations, but are identified through Lagrangian trajectories so that they observe the same air mass. By this approach, dynamic effects in ozone changes can be neglected and only chemical changes remain.

Sasano *et al.* [2000] applied the similar technique to ozone profile data obtained with a satellite-borne sensor, ILAS, and presented preliminary estimates of the ozone loss rates and amounts. Since ILAS provides data not only on ozone but also on other chemical species simultaneously obtained at the same time and at the same location, further comprehensive analysis is possible. This paper presents results of reanalysis on the ozone loss rates and amounts, and focuses on changes in nitric acid ( $\text{HNO}_3$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ) estimated with the same 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 (ozone,  $\text{HNO}_3$ ,  $\text{N}_2\text{O}$ ,  $\text{NO}_2$ ,  $\text{CH}_4$ ,  $\text{H}_2\text{O}$ ) by infrared spectrometry and aerosol extinction coefficient by visible [Sasano *et al.*, 1999, and references therein]. The present analysis uses the ozone,  $\text{HNO}_3$ , and  $\text{N}_2\text{O}$  data from Version 4.20 products, among which validation of ozone products are discussed by Sugita *et al.* in this symposium. The UKMO assimilation data are used for meteorological analyses and trajectory calculations.

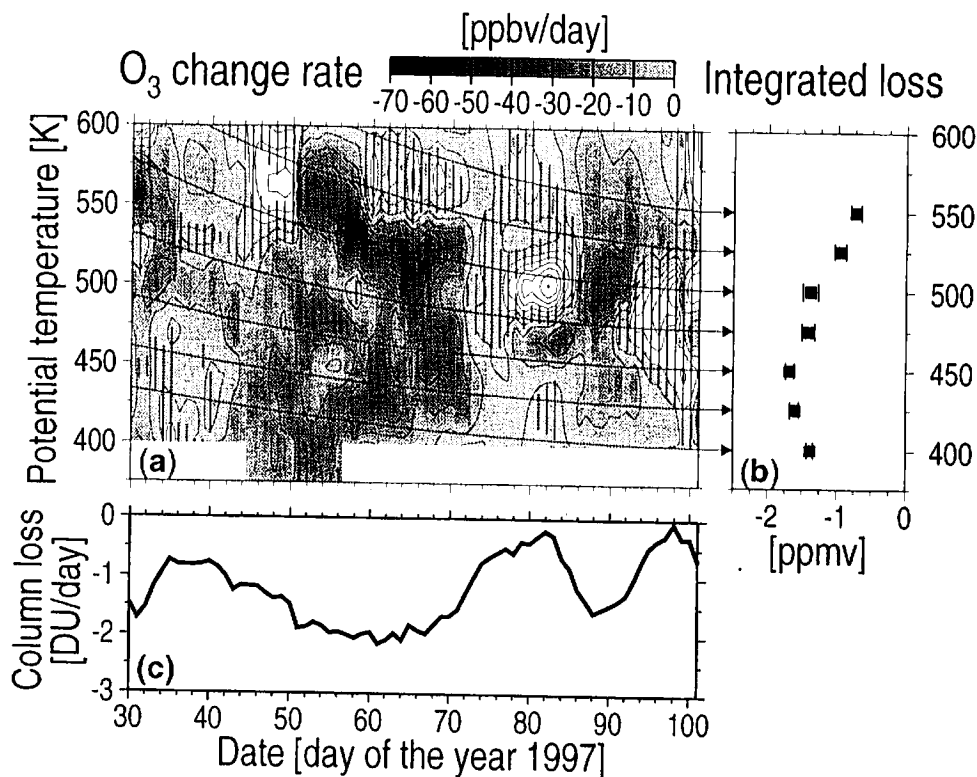
An air parcel that the ILAS sounded twice at different locations and at different times (a double-sounded air parcel) are searched from the ILAS data set with 10-

day forward isentropic trajectories, and an ozone change rate was calculated from the two profiles for each air parcel with diabatic descent effects taken into consideration. Trajectories are calculated on 11 potential temperature levels (from 350 K to 550 K with 25 K intervals, 600 K, and 700 K) using the UKMO trajectory calculation tool. The criterion for determining whether an air parcel starting at one ILAS measurement point comes close enough to another ("Match radius" in Rex *et al.*) was decided as 400 km. To reduce an error in calculation of trajectories, trajectories for which the difference between the maximum and the minimum potential vorticity along the trajectory exceeds 15% of the mean value are discarded. The diabatic cooling rates averaged over the polar vortex were taken from Knudsen *et al.* [1998]. The ozone mixing ratio at the second ILAS measurement is modified by potential temperature changes due to diabatic cooling during the flight time. Since chemical destruction of ozone is considered to occur mainly inside a polar vortex, we focus only inside the polar vortex. See Sasano *et al.* [2000] for details of the analysis method.

Under these conditions, we obtained 2,993 double-sounded air parcels from January to April 1997 between 375 K and 600 K inside the Arctic vortex. A statistical treatment is applied to the subset of measurement pairs to calculate ozone change rates for each day. Each subset consists of matching pairs that are gathered within 7 days before and after each target day. Assuming that ozone changes are linearly proportional to the sunlit time along the trajectory, a proportional coefficient (ozone change rate against sunlit time) is calculated using the least-squares method from each subset of data. 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 same procedure is applied to  $\text{HNO}_3$  and  $\text{N}_2\text{O}$ .

## Results and discussion

Figure 1(a) shows contour plot of ozone change rates (in ppbv per day) as a function of potential temperature and date. Ozone losses took place at almost all the altitudes during February and March. The maximum ozone loss rate was found on the 525 K potential temperature surface in late February, amounting to  $62 \pm 6.7$  ppbv/day. The integrated ozone loss from February to early April 1997 showed its maximum of  $1.7 \pm 0.1$  ppmv at the surface that followed the diabatic descent of the air parcels and reached the 450 K level on April 11 (Figure 1b). This is about 50% of the initial (January 30) ozone concentration. Figure 1(c) shows ozone column



**Figure 1.** (a) Contour plot of ozone change rates (in ppbv per day) as a function of potential temperature and date (day of the year 1997). A contour interval is 10 ppbv/day, and ozone loss regions are shaded. Vertical bars indicate the region with less statistical significance than 99%. Smooth thin curves show potential temperature changes of air parcels (diabatic descent of air masses) [adopted from Knudsen et al., 1998]. (b) Integrated ozone changes from January 30 to April 11 (day 30-101) (day 57-101 and day 39-101 for the uppermost two levels, respectively) along the descent curves of air masses as a function of potential temperature. Error bars represent one sigma. (c) Ozone column change rates (in DU per day) as a function of date, which were obtained by integrating the local ozone change rates (in number density per day) from 400 to 600 K. Monthly-mean temperature and pressure profiles inside the vortex are used to convert the unit of ozone change rate from mixing ratio to molecules per square meters.

change rates (in DU per day) integrated from 400 to 600 K. The accumulated column ozone loss from January 30 to April 11 is  $86 \pm 0.3$  DU.

$\text{HNO}_3$  change rates indicate significant negative trends at almost all the altitudes during February and March. Large  $\text{HNO}_3$  decreases ( $-0.3 \sim -0.4$  ppbv/day) appeared during the period from mid-February to early-March between 400 K and 500 K surfaces (figure not shown). There may be several causes of the changes in  $\text{HNO}_3$  in the air mass, e.g., photolysis, condensation or evaporation on particles, and irreversible loss, known as denitrification. While these large  $\text{HNO}_3$  decreases are likely related to denitrification, further analysis will be needed.

$\text{N}_2\text{O}$  is originally considered as a conservative quantity in a same air mass. If a change in  $\text{N}_2\text{O}$  is significantly large, it may indicate there are some problems in the present analysis.  $\text{N}_2\text{O}$  change rates derived here show relatively small, but significant positive trends at almost all the altitude regions during February and March (figure not shown). There may be several reasons as follows: 1) diabatic descent of air mass may be overestimated, 2)  $\text{N}_2\text{O}$  profiles may have some trends with time, 3) there may be dynamical effects such as small scale diffusion or mixing.

Further careful study is required to get better estimates and understanding of the ozone loss in the Arctic stratosphere.

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