

## **Popular Summary**

### **A review of the Match technique as applied to AASE-2/EASOE and SOLVE/THESEO 2000**

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Over the last decade, European scientists have studied ozone loss in the Arctic winter stratosphere using Match, a clever technique that combines ozonesonde measurements and a model of atmospheric dynamics to investigate chemical ozone loss. Previous Match publications have reported alarmingly large loss rates, particularly in January. These loss rates cannot be explained by the currently accepted polar stratospheric chemistry. We apply the GSFC trajectory model in an attempt to duplicate the Match results during the winters of 1992 and 2000. Although we find loss rates similar to those previously published for most of the two study periods, we are unable to produce the very large loss rates in January 1992. Furthermore, our results indicate larger uncertainties should be associated with the loss rates than those appearing in the original Match publications. We find that the calculated ozone loss rates are extremely sensitive to the precise trajectory paths calculated for each trajectory in the month of January. Integrated ozone loss in both years compare well with those found in numerous other studies including a potential vorticity/potential temperature approach. Finally, we suggest an alternate approach to Match using trajectory mapping that more accurately reflects the true uncertainties associated with Match and reduces the dependence upon filters that may bias the results of the original Match technique by rejecting  $\geq 80\%$  of matched sonde pairs and  $> 99\%$  of matched sonde observations.

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## **A review of the Match technique as applied to AASE-2/EASOE and SOLVE/THESEO 2000**

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**Abstract.** We apply the GSFC trajectory model with a series of ozonesondes to derive ozone loss rates in the lower stratosphere for the AASE-2/EASOE mission (January – March 1992) and for the SOLVE/THESEO 2000 mission (January – March 2000) in an approach similar to Match. Ozone loss rates are computed by comparing the ozone concentrations provided by ozonesondes launched at the beginning and end of the trajectories connecting the launches. We investigate the sensitivity of the Match results on the various parameters used to reject potential matches in the original Match technique and conclude that only a filter based on potential vorticity changes along the calculated back trajectory seems necessary. Our study also demonstrates that calculated ozone loss rates can vary by up to a factor of two depending upon the precise trajectory paths calculated for each trajectory. As a result an additional systematic error might need to be added to the statistical uncertainties published with previous Match results. The sensitivity to the trajectory path is particularly pronounced in the month of January, the month during which the largest ozone loss rate discrepancies between photochemical models and Match are found. For most of the two study periods, our ozone loss rates agree with those previously published. Notable exceptions are found for January 1992 at 475 K and late February/early March 2000 at 450 K, both periods during which we find less loss than the previous studies. Integrated ozone loss rates in both years compare well with those found in numerous other studies and in a potential vorticity/potential temperature approach shown previously and in this paper. Finally, we suggest an alternate approach to Match using trajectory mapping that appears to more accurately reflect the true uncertainties associated with Match and reduces the dependence upon filters that may bias the results of Match through the rejection of  $\geq 80\%$  of the matched sonde pairs and  $>99\%$  of matched observations.

## 1. Introduction

Significant progress has been made in understanding the photochemistry of the polar stratosphere since the ozone hole began to appear in the 1980's [Solomon, 1999]. An important demonstration of our understanding, however, is our ability to reconcile the prediction of photochemical models with observed ozone loss. In the Arctic winter stratosphere this problem is especially challenging because the Arctic vortex is less well isolated than the Antarctic vortex and because in the beginning of winter, ozone amounts inside the vortex are higher than outside for altitudes below about 25 km. Thus separating changes in Arctic ozone due to dynamic processes (such as mixing) from changes due to chemical loss is a challenge.

One approach to untangling dynamic and chemical processes in estimating ozone loss is to use measurements of a conservative trace gas species made at the same time as the measurements of ozone. Each ozone observation is tagged with a simultaneous measurement of the trace gas species. Subsequent ozone measurements are then compared to prior ozone measurements that were tagged with similar values of the conservative trace gas species. Chemical ozone loss can be inferred from shifts in the ozone-conservative trace gas correlation. For example, Schoeberl *et al.* [1991] used simultaneous N<sub>2</sub>O and O<sub>3</sub> measurements to estimate Arctic ozone loss during the late winter as part of the Airborne Arctic Stratospheric Expedition (1989).

Sinnhuber *et al.* [2000] use a passive ozone tracer in their chemical transport model and estimate ozone loss by comparing ozone observations with the value of the passive ozone tracer from the model.

*Plumb et al.* [2000] show that conservative tracer-ozone correlations will evolve even in the absence of chemical processes due to continuous dynamic mixing processes. As a result, conservative tracer-ozone correlations should not be applied over extended periods. Failure to account for such changes in the correlative relationships can lead to incorrect estimations of vortex ozone loss and denitrification. To reduce the probability of mistakenly attributing ozone changes to chemical process rather than such mixing processes, *Richard et al.* [2001] compute the ozone loss during the SOLVE (Sage III Ozone Loss and Validation Experiment) 1999-2000 winter period using ozone and two conservative tracers.

Unfortunately most ozone measurements are made without the simultaneous measurement of long-lived tracer fields (e.g. lidar measurements, some satellite measurements, and ozonesondes). Thus we need to be able to estimate ozone loss without the use of long lived tracers.

Pseudo-tracers have also been used to separate dynamics from chemistry in estimating ozone loss. For example, *Manney et al.* [1994] and *Lait et al.* [2002] use potential vorticity (PV) as a pseudo-tracer to estimate ozone loss, but this technique requires high quality PV computations, and PV is not strictly conserved under diabatic processes.

Another approach to this problem, and the focus of this paper, involves the combination of ozonesonde observations with a simulation of atmospheric dynamics as calculated by a trajectory model in a technique called Match, which has become one of the most widely employed approaches for the calculation of chemical ozone loss rates [*von der Gathen et al.*, 1995; *Rex et al.*, 1997, 1998, 1999, 2002; *Schultz et al.*, 2000, 2001]. By tracking an air mass measured by one ozonesonde through space and time until it arrives

at the location of a second measurement by another ozonesonde, we can infer chemical ozone loss from the observed change in ozone between the two measurements.

*Schoeberl et al.* [2002] introduce a variant on the Match technique that uses many sources of data (sonde, satellite and aircraft) to initialize air parcel trajectories. By comparing new observations with the ozone values associated with the older, advected air parcels, chemical ozone loss can again be inferred.

This paper focuses on the original Match technique, our version of Match, and a new variant on Match using trajectory mapping [*Morris et al.*, 1995]. In previous studies, the Match technique has been applied to data from 1992 – 2003 in the Arctic and 2003 in the Antarctic. Published ozone loss rates during cold Arctic Januaries are generally about 30% larger than can be explained by our current understanding of polar stratospheric chemistry, with one to two individual data points in January 1992 that exceed model values by more than a factor of two. Published results reveal some alarmingly large loss rates for the Arctic, well beyond those that can be explained by our current understanding of polar stratospheric chemistry. Below we will summarize the original Match technique, delineate the differences between the original technique and our version of Match, and describe an alternate approach to Match based upon trajectory mapping. We confine our data analysis to the two years 1992 and 2000, corresponding to the AASE-2/EASOE mission and to the SOLVE/THESEO 2000 mission respectively.

## 2. Methodology

We begin with a brief discussion of the characteristics of ozonesonde data that form the basis of Match. We then review the original Match technique as employed in the series of Match papers (e.g., *Rex et al.*, 1998). Since our first research task is to reproduce

the results achieved by Rex and his collaborators for these two missions, we discuss the precise method we used in our version of Match, highlighting the differences with the original Match technique. Next we motivate and introduce a new version of Match using trajectory mapping that we believe yields more realistic estimates of the uncertainties associated with the Match technique. For comparison, we also provide results from the well-established pseudo-tracer approach using PV and potential temperature (the PV/Theta approach).

## **2.1 Ozonesonde data and filtering:**

The electrochemical concentration cell (ECC) type [Komhyr, 1964, 1969] and Brewer-Mast ozonesondes are a simply designed, lightweight, and inexpensive balloon-born instruments used for measuring the vertical distribution of atmospheric ozone to an altitude of 40 km. Numerous intercomparisons with other ozone measuring instruments [Kerr *et al.*, 1991; Komhyr *et al.*, 1995, Reid *et al.*, 1996] have demonstrated that ozonesondes are generally reliable. During the STOIC 1989 campaign (Komhyr *et al.*, 1995) the ECC sonde precision, when compared to ground based LIDARs, microwave ozone instruments, and ozone photometers, was determined to be  $\pm 5\%$  below to 10 hPa ( $\sim 32$  km) in the stratosphere (the uncertainty in the troposphere was found to be  $\pm 6\%$  near the ground and  $-7$  to  $17\%$  in the upper troposphere and in the stratosphere was found to be  $-14$  to  $6\%$  at 4 hPa or 38km). The overall error in the soundings are thought to originate from four different sources: the background current of the electrolytic cells, the variations in pump efficiency with decreasing pressure, the accuracy of the measurement of the air temperature in the cathode chamber, and the cell's response time to changing ozone con-

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centrations. Thus, there is a need to examine the procedural aspects involved in the sonde preparation and the post-flight data analysis.

For this study, over 700 ozonesondes were launched during the AASE2/EASOE (January 1992 – March 1992) and over 700 more were launched during the SOLVE/THESEO 2000 (November 1999 – March 2000) polar campaigns. Soundings from 26 (AASE2/EAOS, not shown) and the 32 (SOLVE/THESEO 2000) stations depicted in Figure 1, have been homogenized using multiple quality control criteria. All ozonesonde data were obtained from the campaign CD-ROMS and the World Meteorological Organization's (WMO's) World Ozone and Ultraviolet Data Centre (WOUDC, [http:// www.msc-smc.ec.gc.ca/woudc/](http://www.msc-smc.ec.gc.ca/woudc/)).

The first filtering process we apply in our version of Match, which is similar to the approach employed by *Bojkov and Bojkov* [1997], focuses primarily on the control of the flight data. The filter ensures that each record from an ozonesonde sounding includes a pressure and a temperature measurement, that the altitude gap between ozone measurement is not larger than 500 m (~90 s data gap), and that the sounding reaches an altitude with a pressure of ~20 hPa (23 – 25 km). In addition, this initial filtering process also checks for and removes telemetry and ozone “spikes”, and flags ozone partial pressure measurements under 1 mPa.

The second filter pass applied to the remaining dataset in our version of Match involves the visual analysis of the measured ozonesonde box temperature. Since, the ozone amount in the sampled air is derived from [*Komhyr and Harris, 1971*]

$$p_{03} = 4.307 \cdot 10^{-4} * I * T_p * t$$

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where  $p_{O_3}$  is the partial pressure of ozone (mPa),  $i$  is the sensor current due to ozone ( $\mu A$ ),  $T_p$  is the sonde box temperature (K), and  $t$  is the time in seconds taken by the ozonesonde gas-sampling pump to force 100 mL of air through the sensor. Therefore, a 3K error in the measured box temperature translates into a one percent error in the sampled ozone measurement. In practice, about 2% of the sondes show unusual behavior in the recorded box temperature data.

From the initial set of 3677 possible sonde-to-sonde matches in AASE2/EASOE on the 475 K surface and 3423 possible sonde-to-sonde matches in SOLVE/THESEO 2000 on the 450 K surface, 3071 and 2813 were left respectively after exclusively applying these two filters in our version of Match. As a result, these two filters eliminate 15 – 20% of the matches. The reader should keep in mind that differences remain in this homogenized dataset due to the varied operational procedures employed by the stations – among the uncertainties are the differences in instrumentation, the pump efficiency curves employed for the soundings analysis, the box temperature measurements, and the pre-launch ozonesonde calibration procedures.

For the original Match technique, the elimination of individual ozonesondes is performed by the Finnish Meteorological Institute in Sodankylä and Alfred Wigner Institute (AWI) in Potsdam. In practice ~10% of the data are eliminated by these filters in the original Match technique.

## 2.2 The Original Match Technique

Match campaigns since 1994 involve a coordinated launch of ozonesondes based upon predictions derived from running a trajectory model that uses forecast winds from the European Centre for Medium-range Weather Forecasts (ECMWF). The AASE-



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2/EASOE campaign did not coordinate the launch of ozonesondes for Match. As a result, the procedure for Match in 1992 begins with trajectories calculated in the analysis mode (below).

In Match, each ozonesonde launch triggers the initialization of air parcels in a trajectory model [*Peterson and Uccellini*, 1979] along the sonde profile geographically coincident with the sonde location. In forecast mode, isentropic trajectories are run using ECMWF forecast wind fields ( $2.5^0 \times 2.5^0 \times 6$  hours) [*Rex et al.*, 1999]. A diabatic correction to the isentropic trajectories is applied using *Lacis and Hansen* [1974] for short wave heating and *Dickinson* [1973] for infrared cooling. For all years except 1992, the model is first run in forecast mode to coordinate ozonesonde launches and thereby improve the prospects of a match occurring. When these forecast trajectories closely approach (within 350 km) another ozonesonde launch facility, a second, matching ozonesonde is launched [*Rex et al.*, 1999].

After the launch of the second ozonesonde, a new set of trajectories are calculated, this time with the trajectory model running in an analysis mode, in other words, using as input the analyzed wind fields from ECMWF ( $1.5^0 \times 1.5^0 \times 6$  hours) [*Rex et al.*, 1998]. The trajectories are integrated forward in time in a diabatic mode with heating rates derived from the Universities' Global Atmospheric Modeling Program (UGAMP) General Circulation Model (GCM) as established by *Geleyn and Hollingsworth* [1979] for AASE-2/EASOE [*Rex et al.*, 1998] and from radiative transfer scheme of the SLIMCAT 3-D chemical transport model [*Chipperfield*, 1999] for SOLVE/THESEO 2000 [*Rex et al.*, 2002]. We note that for 1992, the ECMWF winds are output on 19 levels from 1000 hPa to 10 hPa, while in 2000, ECMWF winds are output on 60 levels and 0.1 hPa. Although

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we have not conducted an appropriate sensitivity study, such differences in the vertical resolution of the ECMWF winds might have an impact on the Match results. The analysis trajectories are limited to 10 days duration and help in the determination of whether or not an actual match occurred. In practice ~80% of forecast matches result in confirmed matches.

Several quality control measures insure the integrity of each match. Ozonesonde profiles are interpolated to the altitude at which the match occurs. Interpolations are not performed, however, onto surfaces that lie within vertical gaps in the ozonesonde profile that exceed 500 m [Rex *et al.*, 1998, 1999]. Given typical ascent rates, this distance implies a temporal gap of approximately 90 seconds in the ozone profile. Station-to-station and year-to-year differences in the time averaging of ozonesonde profiles could result in inconsistencies in the impact of this criterion on Match.

As the ozonesonde ascends, its latitude and longitude coordinates vary due to transport by the local winds. Separate instrumentation on the same balloon payload records the winds thereby permitting the computation of latitude and longitude as a function of potential temperature surface for the purposes of initializing each air parcel within the trajectory model. For those sondes which do not record local winds, the winds are interpolated from the 3-D grid of the analyzed wind fields from ECMWF to the ozonesonde profile so that a calculation similar to the one described above can be performed.

To track each air parcel along the measured profile, a tight cluster of 7 parcels is initialized in the trajectory model on each potential temperature (Theta) surface with a valid measurement: 5 of these parcels are on the Theta surface of interest; one is 5 K in Theta directly above and one 5 K directly below. The center of the cluster of 5 on the same

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theta surface is at the ozonesonde location. The other 4 in that cluster of 5 are located 100 km away, one each north, south, east, and west [Rex *et al.*, 1999]. The Match study for 1991/1992 was the exception in that this cluster approach was not employed. In all cases, Match trajectories are limited to 10 days duration.

In determining a valid match, only the central parcel in the cluster of 7 is used. If the central parcel lies within the specified Match radius of the location of the new ozonesonde observation, the corresponding ozone observations are said to have been made within the same air mass, and a match is said to have occurred. In *von der Gathen* [1995] the Match radius used for AASE-2/EASOE is 500 km. In *Rex et al.* [1998], the Match radius used for AASE-2/EASOE is 475 km (1992) while in *Rex et al.* [2002], the Match radius is 400 km (2000). In each case, Rex found the Match radius that achieved a minimum in statistical uncertainty of the ozone loss rate calculation. *Rex et al.* [2002] are able to use a tighter Match radius for SOLVE/THESEO 2000 since launches in 2000 are coordinated using Match forecast trajectories whereas in 1992 for AASE-2/EASOE, Match is only run in analysis mode. Near the vortex boundary, the shape of this Match region is altered from a circle (used for AASE-2/EASOE) to an oval (used since SOLVE/THESEO 2000) with a major axis of 500 km parallel to lines of constant PV and a minor axis of 300 km in the perpendicular direction [Rex *et al.*, 1999]. Again, the changes were implemented in the original Match technique in an attempt to minimize the statistical uncertainty associated with the Match results.

The 6 other parcels in each cluster are used to diagnose the validity of the corresponding central trajectory and to filter out air masses that are more likely to have been influenced by mixing processes. Clusters of parcels that remain spatially close together are

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more likely to describe actual air parcel trajectories. At the time of the match (second ozonesonde measurement), the distance from the central parcel to each of the other 6 parcels in the cluster is calculated. If that distance exceeds 1200 km for the 5 parcels that began on the same potential temperature surface or 1300 km for the 2 parcels that began 5 K above or below, then the match is discarded.

A limit on the vertical gradient in ozone concentrations measured by the ozonesondes is also imposed. For Match during AASE-2/EASOE, ozone is allowed to vary by 15% over the altitude range 2 K above to 2 K below the potential temperature surface of the match and 25% over the altitude range of 5 K above to 5 K below the surface of the match [Rex *et al.* 1998]. For Match during SOLVE/THESEO 2000, these restrictions are 20% and 30%, respectively [Rex *et al.*, 1999]. Such restrictions serve two purposes. First, ozonesonde profiles within the Arctic polar vortex often contain sharp gradients due to imbedded filaments of extra-vortex air. Such filaments do not characterize the vortex air mass and, therefore, can complicate the interpretation of the Match results. Second, by examining only those parts of the profile with small vertical gradients in ozone, uncertainties in the diabatic portion of the trajectory calculation that might bias ozone loss rate calculations are reduced.

Each tracked air parcel is only permitted a single match with each other sonde on a given day, although it may match multiple sondes on the same day. Furthermore, air parcel trajectories that exhibit significant deviations in PV are deemed to be unreliable. Therefore, the potential vorticity as calculated along the air parcel trajectory is not allowed to vary by more than 40% between its maximum and minimum values for AASE-2/EASOE [Rex *et al.*, 1998] and 25% for SOLVE/THESEO 2000 [Rex *et al.*, 2002].

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ECMWF analyses switched from a 3-D to a 4-D assimilation process, greatly reducing the noise in the PV fields from the 1992 data to the 2000 data. The  $\Delta PV$  limits are suggested by *Rex et al.* [1999] at the point where they observed an increase in the scatter of the matched ozonesonde observations. As with the case of the Match radius, the change in the  $\Delta PV$  criterion also is related to the fact that ozonesonde launches were not coordinated in 1992 for AASE-2/EASOE, reducing the number of available matches. For later campaigns during which the launches are coordinated, more restrictive criteria could be enforced while still resulting in a sufficient number of matches from which to calculate ozone loss rates.

Only matches that occur within the polar vortex or near the edge of the vortex are included in the original Match studies. *Rex et al.* [1998] use a derived quantity that they call “normalized PV” to locate the vortex edge. This quantity is based upon the scaled PV of *Dunkerton and Delisi* [1986] and is defined so that the normalized PV and Ertel’s PV have the same values at the 475 K isentropic surface. *Rex et al.* [1998] use a vortex boundary of 36 normalized PV units ( $1 \text{ PVU} = 10^6 \text{ K m}^2 / \text{s} / \text{kg}$ ), so that the Match data include air parcels at the vortex edge. As a result, the vortex size is 10 – 15% larger than the area poleward of the PV contour at the maximum PV gradient.

To compute the ozone loss rate (ppbv per sunlit hour), the total amount of sunlight along the back trajectory is calculated. To determine the number of hours of solar illumination, a careful calculation is performed to determine if the center of the solar disk is visible at the precise location of the air parcel. This calculation includes atmospheric refraction effects and the non-spherical shape of the Earth. The time over which the air parcel can see the center of the solar disk is integrated to compute hours of solar illumina-

tion. The ozone loss rate can be determined by dividing the difference between the ozone measurements of the new ozonesonde and that of the original ozonesonde by the total number of hours of solar illumination.

A more robust approach than calculating a loss rate for each match is to calculate the loss rate for an ensemble of matches. In practice, matches are accumulated over a 14-day (1992) or a 20-day (2000) period. A regression is performed of ozone change on hours of solar illumination to produce a line-of-best-fit. The slope of that line is the ozone loss rate. The ozone loss rates are computed this way once per week. Uncertainty in the calculated ozone loss rates is computed using the standard statistical methods. The regression line is forced to pass through the point (0,0) since air parcels that have not been exposed to sunlight should not experience chemical ozone loss. *Rex et al.* [1998] and *Rex et al.* [2003] performed multi-variable regression on sunlit hours and dark hours using Match data and observed little to no ozone change during the dark hours.

### 2.3 Our Version of the Match Technique

In our attempts to reproduce the results of *Rex et al.* [1998, 2002], we have used a very similar approach to that described above with the following exceptions. We initialize parcels at every altitude for which the ozonesonde data files report a measurement. Each observation is initialized as a cluster of parcels, one each 50 km north, south, east and west of a central parcel at the location of the ozonesonde measurement, but only the center one is used to define a match. We include matches within the polar vortex as defined using modified potential vorticity (MPV) [*Lait*, 1994] and a maximum gradient definition of the vortex boundary [*Nash et al.*, 1996]. To approximate the weak definition of the vortex boundary used in the original Match technique, we use a MPV criterion

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of the weakest edge (defined by the nearest to the vortex boundary of the maximum of the second derivative of the MPV).

To determine the number of hours of solar illumination, the parcel location and local time at each point along the trajectory is used to compute the solar zenith angle (SZA). The parcel is considered to be illuminated if the SZA is less than  $95^{\circ}$ . At 20 km, a height very near that of all of the potential temperature surfaces considered in this study, this SZA corresponds to the sun on the horizon. We note that although the photochemistry may initiate at a SZA slightly greater than  $95^{\circ}$ , the uncertainty in the trajectories themselves will produce larger errors in the end result than that incurred by this SZA error.

Our version of Match permits unlimited matches for each sonde every day, however restricts air parcels from each sonde to match any other given sonde exactly once. To provide an estimate of the robustness of the results, we introduce a random component to select subsets of matches with which to compute ozone loss rates. This random selection process is done in an iterative way so that a wide range of possible outcomes are represented.

To compute uncertainties, we examine both the scatter of these outcomes as well as a boot-strap approach [Efron, 1982] applied to any one particular outcome. In the boot-strap approach, a random subset of size equal to the size of the original data set is chosen. The subset is allowed to include duplicates. Linear regression is performed on the subset; the process repeated; and the slopes accumulated. The uncertainty of the ozone loss rate can be estimated by the standard deviation of these slopes, so long as the scatter in the variance of the change in ozone between the two ozonesondes is not correlated with the amount of solar illumination.

To demonstrate that this condition is satisfied in our data, Figure 2 shows that the standard deviation of the change in ozone as calculated for the SOLVE/THESEO 2000 mission from January – March 2000 is independent of the amount of sunlight the air parcel receives. The data in this figure represents the combined results from the 450 K (~19 km) and 500 K (~23 km) surfaces. Thus, the boot-strap method appears to be a justifiable and reliable approach to estimating the uncertainties in the ozone loss rates for Match.

We compute ozone loss rates daily so that it is easy to identify days for which something unusual occurs. Finally, we employ wind fields ( $3.75^{\circ} \times 2.5^{\circ} \times 24$  hours) from the United Kingdom Meteorological Office (UKMO) [Swinbank and O’Niell, 1994] with heating rates calculated as in Rosenfield *et al.* [1994] and use trajectories of up to 14 days duration as calculated by the Goddard Trajectory Model [Schoeberl and Sparling, 1995]. Lucic *et al.* [1999] note that the UGAMP derived heating rates used by Rex *et al.* [1998] result in about 0.2 K/day more descent than those computed by Rosenfield *et al.* [1994] for the 1991/1992 winter season, a consideration to keep in mind when comparing our results with those from the original Match technique. Also worth considering is the impact of the reduced time resolution of our wind fields compared to those utilized in the original match study. Waugh and Plumb (1994) noted that trajectory calculations depend strongly on the time resolution of the meteorological fields.

## 2.4 Potential Vorticity/Potential Temperature Approach

As a check on our results, we include the calculation of integrated loss rates over the winters of 1992 and 2000 using the PV/Theta approach. The PV/Theta approach, built upon the ideas of McIntyre [1980], originally put forward by Schoeberl *et al.* [1989], and



finally formalized by *Schoeberl and Lait* [1992], takes advantage of the quasi-conserved nature of both PV and Theta and has been applied in numerous studies to problems involving sparse data sets [*Lait et al.*, 2002; *Randall et al.*, 2002; *Strahan*, 1999a; *Strahan et al.*, 1999b; *Lucic, et al.*, 1999; *Manney et al.*, 1999; *Plumb, et al.*, 1995, *Redaelli, et al.*, 1994; *Salawitch, et al.*, 1993; *Lait, et al.*, 1990; *Douglass et al.*, 1990; *Salawitch, et al.*, 1990]. Ozone observations are located in a PV/Theta coordinate space using values of PV and Theta which have been corrected for diabatic effects by means of trajectory calculations. For each point in a regularly-spaced grid in the PV/Theta coordinate space, a weighted linear least-squares fit is applied to the ozone data near that gridpoint to obtain a chemical loss rate.

With regards to this study, we used all available ozonesondes north of  $60^{\circ}$  latitude associated with PV values among the highest 10% of all PV values on a given Theta surface on a given day. These sonde data were used to construct the PV/Theta/ozone relationships. As a result, the data input in the PV/Theta analysis represent only data at the core of the polar vortex.

Trajectory calculations are used to determine descent and to identify parcels that crossed the vortex boundary. Those parcels for which trajectory calculations indicated a variation in the Modified Ertel's Potential Vorticity (MPV) of more than 12.5% are eliminated from consideration. The trajectories are calculated to the date at the middle of the analysis range. For example, when studying data from January 1 through February 29, 1992, trajectories are run either forward or backward in time, as appropriate, to January 30<sup>th</sup>. As a result, comparing loss rates at a particular Theta surface is most accurate

for January 30<sup>th</sup>. PV/Theta loss rates will be somewhat displaced vertically as compared to Match results for earlier or later days.

### 3. Diagnostics and Sensitivity Studies

In this section, we demonstrate the equivalence of our version of Match with the original version. We also explore the sensitivity of our version of Match to the Match filters applied by *Rex et al.* [1998, 2002] and described above. As we show below, most of the filters seem not to impact the results.

First, we attempt to reproduce Figure 6 from *Rex et al.* [1998]. In that figure, the authors show the linear relationship between change in ozone and the hours of sunlight illumination as computed from data for the period of the largest ozone loss rates, January 4 – February 9, 1992, during the AASE-2/EASOE campaign. We therefore bin all of the Match data from this period by the number of hours of solar illumination with bins 20 hours wide, plotting one data point for each 10 hours of sunlight illumination, as did the original authors.

Figure 3 shows both the original data from *Rex et al.* [1998] (red) and data from our version of Match (blue). Note that we extend the data beyond the limit of 65 hours of sunlight illumination that appears in the original figure to near 85 hours of sunlight. Error bars for both sets of data represent one standard deviation. Our data points represent the average of both the ozone change and the number of hours of sunlight illumination within each bin and are plotted with an associated error bar for both quantities based on the variance within each bin.

Based on Figure 3, a powerful case can be made that the ozone change is linearly related to the amount of sunlight to which the air parcel is exposed, as is expected from our

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current understanding of polar winter photochemistry [Solomon, 1999]. Furthermore, the extremely good agreement between the results of *Rex et al.* [1998] and our data indicates that we have done a reasonably good job of reproducing Match. We do note, however, that despite the fact that the error bars for both sets of results overlap, our data systematically appear to indicate less loss per hour of sunlight than the original data of *Rex et al.* [1998]. This difference may be associated with differences in our transport and/or radiation schemes.

Next, we examine the sensitivity of the ozone loss rate results to the following parameters: (1) PV difference along the back trajectory; (2) spreading of the cluster of parcels initialized for each ozonesonde measurement; (3) the duration of trajectories between Matches; (4) the precise SZA at which the terminator is defined in the calculation of the number of hours of solar illumination; and (5) the combined use of all the filters in the original Match technique versus the use of far fewer filters. From these sensitivity studies we find that the PV difference along the back trajectory appears justifiable and that the definition of the day/night terminator can impact the loss rate calculations, especially in January. From our study, it would appear that the remainder of the filters do not significantly impact the ozone loss rate calculations.

### 3.1 PV Differences

To examine the impact of the PV difference along the back trajectories on the ozone loss rate, we bin all the data from 1992 and 2000 by the ozone loss rate and plot the mean loss rate and standard deviation for each year in Figure 4. In the figure, data from AASE-2/EASOE appears blue while data from SOLVE/THESEO 2000 appears red. The thick lines represent the mean quantities while the thin lines represent the mean plus and minus

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one standard deviation. The solid black line at 40% represents the filter value employed by *Rex et al.* [1998].

Figure 4 shows that the average loss rates are well behaved for PV differences of less than 40%. In other words, the standard deviation of the mean ozone loss rate remains relatively constant over this domain. For SOLVE/THESEO 2000, not only is the standard deviation constant, but the ozone loss rate remains nearly constant over the entire domain as well. Beyond 50% PV differences, it is clear that neither the ozone loss rate nor the standard deviation remain constant or predictable. Therefore, the cut-off value of 40% used by *Rex et al.* [1998] appears to be a valid and useful parameter by which to filter out less reliable Match data.

*Grooß and Müller* [2003] performed a sensitivity analysis of the original Match technique to the PV filtering criterion using the Chemical Lagrangian Model of the Stratosphere (CLaMS). They applied a cut-off value for PV of 25% as in *Rex et al.* [1999] and used for all Match campaigns after AASE-2/EASOE. After applying the PV filter, they observe that the ozone loss rate bias that results from the original Match technique as compared to CLaMS changes from  $+2.40 \pm 0.07$  ppbv/sunlit hour to  $-0.41 \pm 0.08$  ppbv/sunlit hour, a significant effect. We note that in their study, the Match radius is 300 km and the trajectory length is 4 days, parameters different from the original Match technique and our version of Match. Nevertheless, our results concur with those of *Grooß and Müller* [2003], both indicating that the PV filter criterion is an important one for the successful application of Match. From our results a  $\Delta$ PV cut-off of 40% may be optimal both in 1992 and in 2000, so unlike *Rex et al.* [2002], we do not recommend changing the cutoff to 25% for the SOLVE/THESEO 2000 study period.

### 3.2 Cluster Spreading

Next, we examine the sensitivity of the results to the spreading of the cluster of trajectories that was initialized for each ozonesonde observation. Recall that *Rex* [1993] established a criteria to filter Match data for which the trajectory of at least one member of the cluster led to a separation of more than 1200 km from the central parcel at the time of the Match. Figure 5 shows the average ozone loss rate as a function of the maximum spreading of each cluster of trajectories. Again, AASE-2/EASOE is shown in blue while SOLVE/THESEO 2000 is shown in red and the thick lines represent the mean values while the thin lines represent the means plus and minus one standard deviation.

This quantity is much better behaved than the PV differences seen in Figure 4. In fact, it is difficult to assign an appropriate distance at which a transition occurs to justify establishing a cut-off value for parcel spreading on which to filter Matches. Based on Figure 5, it appears that the cut-off for parcel spreading need be no more restrictive than 3000 km and in fact may be entirely unnecessary.

Our results differ from those achieved by *Groob and Muller* [2003]. They found that by applying the cluster spreading filter criterion of *Rex et al.* [1998], the bias in the ozone loss rate calculated in Match compared to CLaMS changes from  $+2.40 \pm 0.07$  ppbv/sunlit hour to  $-0.23 \pm 0.07$  ppbv/sunlit hour, another significant effect. Again, we note that the parameters used by *Groob and Muller* [2003] differ somewhat from those of the original Match technique and our version of Match. Most importantly, *Groob and Muller* [2003] separate parcels in their cluster by 100 km rather than 50 km we use in our version of Match.

### 3.3 Trajectory Duration

We next examine the effect of the duration of Match trajectories on the ozone loss rate calculations. Figure 6 shows the impact of including trajectories of durations of up to 14 days on the resulting ozone loss rates. As in Figures 4 and 5, the blue lines represent AASE-2/EASOE data while the red lines represent SOLVE/THESEO 2000 data. The thick lines represent the mean values while the thin lines represent the means plus and minus one standard deviation.

As can be seen in Figure 6, the shortest duration trajectories show the largest variation in ozone loss rates, both by the fluctuation of the mean and the large uncertainties. This result is not particularly surprising given the fact that the shortest duration trajectories will be associated with the smallest exposures of the air parcels to sunlight. Since the sunlight exposure appears in the denominator of the ozone loss rate calculation, small absolute changes in these small numbers can lead to large changes in the resulting quotient.

Figure 6 indicates that no penalty is incurred with regards to the ozone loss rate calculations by including trajectories of durations of up to 14 days. In fact, the Figure 6 indicates that the uncertainties actually decrease by including these longer duration trajectories. Such a result suggests that the increased error incurred by including longer, and hence more uncertain trajectories, is more than offset by the increased number of matches that result from considering more and longer trajectories. We note that although 14-day trajectory calculations appear at the upper end of the range of trajectory durations recommended in previous trajectory studies [*e.g.*, *Morris et al.*, 1995, *Morris et al.*, 2000] we nevertheless recommend extending trajectory calculations to 14 days for future match analysis.

### 3.4 Solar Zenith Angle of Day/Night Boundary

Next, we examine the impact of the SZA definition for the terminator. An examination of the sensitivity of the ozone loss rates to this quantity is relevant for more reasons than the precise SZA at which the chemistry turns on and off. High sensitivity to this quantity suggests that the precise trajectory path will affect the calculated ozone loss rate. Numerous analyses of trajectory modeling have indicated that while the trajectory path computed for any individual trajectory is not reliable (particularly for calculations of duration greater than a few days), the results from an ensemble of trajectories provides useful and reliable information [e.g., *Morris et al.*, 1995].

While a large number of trajectories is initialized in the Match technique, only a fraction actually are used to compute the ozone loss rates due to the numerous filters employed by and recommended by *Rex et al.* [1998, 1999]. According to its developers, the filters of the original Match technique eliminate 30 – 50% of the matches (Rex, personal communication). We find, however, that  $\geq 80\%$  of potentially matched sonde pairs and  $>99\%$  of potentially matched sonde observations are disregarded in the ozone loss calculations. If those trajectories that survive the filtering are inherently biased with regards to their position relative to the local solar terminator, a bias in the amount of solar illumination may result in a bias in the calculated ozone loss rates.

Figure 7 shows the sensitivity of the ozone loss rate to the definition of the terminator for data from the AASE-2/EASOE time period. The solid colored lines represent loss rates calculated with our version of Match using a range of the SZA criterion from  $90^\circ$  to  $96^\circ$ . (Note that the Figure 7 indicates little difference between the ozone loss rates computed using a  $94^\circ$  SZA criterion and  $96^\circ$  SZA criterion. Examining the difference in trajectories with SZA of  $90^\circ$  versus SZA of  $94^\circ$  lead to trajectory errors of about  $4^\circ$  in

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latitude or  $15^{\circ}$  in longitude for conditions in mid-January at  $65^{\circ}\text{N}$  latitude. Trajectories near the vortex edge where wind speed gradients are large are more likely to experience such errors.) The red squares and associated error bars are again data from *Rex et al.* [1998]. *Rex et al.* [1998, 2002] use a careful calculation of the exact SZA at which the sun disappears below the horizon at each air parcel altitude. In practice, that number varies very little from the  $95^{\circ}$  SZA that we employ in our version of Match. We also recall that the uncertainty in the trajectories themselves likely will result in larger errors than those resulting from the use of  $95^{\circ}$  as the SZA for the terminator.

We can see in Figure 7 that after about day 40 (February 9<sup>th</sup>), the precise definition of this boundary has little impact on the calculated ozone loss rates, with variations between the ozone loss rates at  $\text{SZA} = 90^{\circ}$  and that at  $\text{SZA} = 96^{\circ}$  of only about 1 ppbv/sunlit hour. Before day 35 (February 4<sup>th</sup>), however, we see large differences in the loss rate depending upon the precise SZA chosen, with the largest differences ( $\sim 6$  ppbv/sunlit hour) occurring in early January.

The fact that the calculated ozone loss rates show the greatest sensitivity to the SZA employed in January is not surprising. During January, the number of hours of solar illumination are quite small (and often zero) at the highest northern latitudes. By the middle of March, most of the same latitudes are receiving nearly 12 hours of sunlight per day. As a result, the percent uncertainty in the amount of solar illumination is much greater for a given trajectory in January than in March.

It is also not surprising that the largest discrepancies in the ozone loss rates calculated by *Rex et al.* [1998, 2002] and those presented in this paper appear in January. Slight systematic differences in the trajectory calculations between the ECMWF winds used by



*Rex et al.* [1998, 2002] and the UKMO winds used in this paper could easily lead to differences in calculated ozone loss rates of 4 – 6 ppbv/sunlit hour in January according to Figure 7. In fact, we see that the largest published ozone loss rate from *Rex et al.* [1998] for late January 1992 falls near the curve computed using a day/night terminator with a SZA of  $90^{\circ}$ , although such a terminator is unrealistic for the relevant ozone chemistry and at the altitudes of our study.

We are led to the conclusion from Figure 7 and from the small fraction of trajectories actually selected for inclusion in the Match ozone loss rate calculations that the actual errors associated with the ozone loss rates calculated using Match are much larger than the statistical error bars appearing in previous publications, especially for data in January. Were not so many filters applied to the Match data, the likelihood of an unintentionally introduced selection bias in SZA would be substantially reduced, but the original Match technique and our version of Match include many filtering criteria, which when combined result in the selection of only a small fraction of the Match data as qualifying events. Figure 7 gives us cause for concern in interpreting Match results in January, particularly as related to the extremely large loss rates published by *Rex et al.* [1998] for AASE-2/EASOE during January 1992.

### **3.5 Sensitivity to Population Selection**

We examine Match results after removing all the Match filters except for the Match radius and the definition of the vortex boundary using MPV. We find that the ozone loss rates so calculated fall well within the associated uncertainties as compared to those calculated with our version of Match using all the filters (not shown). Furthermore, the associated error bars for the ozone loss rates are comparable if not smaller than those

associated with the ozone loss rates determined in our version of Match (see discussion in Results section below).

Such results suggest that the five-fold increase in the number of matches which results from elimination of the Match filters more than offset the added uncertainty from the inclusion of more dubious matches in the ozone loss rate calculations. Furthermore, it is comforting to include so many matches and achieve similar results. By not applying the Match filters, we can be sure that we have not accidentally thrown out some good data with the bad nor have we unintentionally biased our results. It may be reasonable to conclude that Match could be as (if not more) effective as the original Match technique by eliminating most if not all of its current data filters.

## **4. Results**

In this section, we present results from our version of Match and from the PV/Theta analysis. Our version of Match yields loss rates of similar magnitude to those published by *Rex et al.* [1998, 2002], although we are unable to reproduce the largest loss rates in January 1992 on the 475 K surface without significantly and unrealistically altering the SZA for the terminator (see discussion in Section 3.4 above). We also find somewhat smaller loss rates in March 2000 on the 450 K and 500 K surfaces than those shown by *Rex et al.* [2002]. Our loss rates do agree well with numerous other studies including model simulations, as we outline below. Finally, our error bars are generally larger in magnitude than those of *Rex et al.* [1998, 2002]. We discuss in detail our approach to the error calculation below.

### **4.1 Results from Our Version of Match**

Figure 8 shows the ozone loss rate as a function of time for the SOLVE/THESEO 2000 period. Each black dot in the figure represents one possible outcome for the ozone loss rate calculation. For example, Figure 9 shows the Match data from the 20-day period January 12 – February 1, 2000. We randomly pick half of these matches from which to compute the line-of-best fit. The randomly selected half appears as the solid red squares while the unused data are open black squares. For the red data points, we find a line-of-best-fit with a slope of  $-4.61 \pm 1.75$  ppbv/sunlit hour. Note that this loss rate is substantially different from that calculated by using all the data of  $-2.74 \pm 1.10$  ppbv/sunlit hour).

The statistical errors associated with the original Match data are similar in magnitude to the scatter the loss rate calculations based on the subsets of data. However, the total uncertainty in the loss rates is larger than that estimated by the standard regression routine (quoted with the slopes above). Furthermore, because the regression is forced through (0,0), the uncertainty estimate associated with the slope will necessarily be reduced as compared to the uncertainty in the estimate of the slope when the line-of-best fit has two free parameters (slope and intercept) as calculated with the standard routines.

Our error analysis approach is illustrated by Figure 9. Subsets of data are randomly selected for each day ( $\pm 14$  days for AASE-2/EASOE or  $\pm 20$  days for SOLVE/THESEO 2000). These subsets permit us to explore the range of possible ozone loss rates. Figure 9 represents one possible outcome. The slope computed using the data highlighted by the red squares in Figure 9 leads to one black dot in Figure 8. The random subsets are generated 200 times for each day. Each of the black dots in Figure 8 therefore represents the loss rate as computed from one such subset. The mean results (the average

of the black dots) is indicated by the thick blue line. The thin blue lines represent the average plus and minus one standard deviation as computed using the scatter of the black dots.

The thick gold lines in Figure 8 represent the average result (thick blue line) plus and minus one standard deviation as computed using the boot-strap technique (described above) to exactly one realization of the random subsets of data (e.g., the subset highlighted in red in Figure 9). The gold lines therefore represent a different and independent estimate of the uncertainty in the data as compared with the thin blue lines which are generated from the scatter of the ensemble of results. As expected, the uncertainty from the bootstrap technique has a similar magnitude to that computed from the scatter of the data.

The solid red squares in Figure 8 represent the loss rates as calculated by *Rex et al.* [2002], and their associated error bars are one standard deviation from the mean as computed using standard regression error algorithms.

For the 500 K Theta surface during SOLVE/THESEO 2000, we find reasonably good agreement between the magnitudes of the loss rates published by *Rex et al.* [2002] and those found in our version of Match, which again closely mimics the original Match in the filters applied to the data. In late January and early February (days 15 – 35) and again in late February and early March (days 56 – 70), our results seem to indicate somewhat smaller loss rates than those of *Rex et al.* [2002]. For the remainder of the time period, however, the loss rates show good agreement. The uncertainty in the scatter of the black dots (represented by the thin blue lines) also shows good agreement with the statistical error estimates of *Rex et al.* [2002] for this level.

Figure 10 shows the results from our version of Match for the 450 K surface during SOLVE/THESEO 2000. While we see generally good agreement in late January through mid February (days 28 – 49), we find that during days 56 – 84, our version of Match produces smaller ozone loss rates than those of *Rex et al.* [2002]. Furthermore, the larger loss rates of *Rex et al.* [2002] in early March 2000 fall outside the error bars of both the *Rex et al.* [2002] data and the boot-strap error estimates for our data. At present, we find no good explanation for the disagreement. It is possible that such differences are indicative of uncertainties inherent in the Match technique for which we have not yet accounted and may be more representative of the true uncertainty of the technique. In any event, we believe the error bars associated with the *Rex et al.* [2002] data during this time period underestimate the true uncertainties associated with these loss rates. Figure 10 clearly indicates that even our statistical error bars are much larger than those of *Rex et al.* [2002] for this level.

Figure 11 shows the results for the 475 K surface during the AASE-2/EASOE period of January through March 1992. We find generally good agreement throughout the period with the notable exception of days 20 – 35 during which *Rex et al.* [1998] report losses of a magnitude never before seen in the Arctic and that are difficult to reproduce with our current understanding of stratospheric chemistry [*Sander et al.*, 2003; *Solomon*, 1999]. Our data show large loss rates during this time period as well, but of half the magnitude. The combined uncertainty of the *Rex et al.* [1998] data and our boot-strap error estimates is larger than the difference in the results. Once again, we have no good explanation for the difference and are led to the conclusion that we both have still underestimated the actual errors inherent in the Match technique. Figure 11 also shows a dis-

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crepancy for the loss rates in mid-February, again with our model showing less loss. By altering the vortex boundary condition, better agreement can be found at this time. The apparent disagreement at this time may be related to the differences between our vortex boundary definition using UKMO meteorological data and that of *Rex et al.* [1998] using ECMWF meteorological data.

#### 4.2 Comparisons with Other Studies

*Newman et al.* [2002] published a summary of results for integrated ozone loss during the SOLVE/THESEO 2000 campaign on the 450 K potential temperature surface. Their Table 8 lists integrated ozone losses over the period January 20 – March 12, 2000 from 14 different studies. Losses ranged from 0.7 ppmv [*Klein et al.*, 2002] to 2.3 ppmv [*Santee et al.*, 2000] and with an average loss of  $1.5 \pm 0.4$  ppmv. *Rex et al.* [2002] use the original Match technique and reports an integrated ozone loss of  $1.7 \pm 0.2$  ppmv over the same time period. Our version of Match yields an integrated ozone loss of  $1.5 \pm 0.6$  ppmv, in extremely good agreement with the other integrated ozone losses listed in *Newman et al.* [2002]. *Lait et al.* [2002] use a PV/Theta approach to estimate ozone loss. For SOLVE/THESEO 2000, they find an integrated ozone loss over this period of  $1.7 \pm 0.3$  ppmv.

No similar compilation has been published for the AASE-2/EASOE campaign for which *Rex et al.* [1998] and *von der Gathen* [1995] published their largest Arctic ozone loss rates. Based on the data of *Rex et al.* [1998], integrated chemical ozone loss for air parcels that descended from 500 K on January 1 to 460 K on February 29, 1992 is  $1.2 \pm 0.3$  ppmv. For the same period, we find an integrated chemical ozone loss at 475 K of  $1.2 \pm 0.4$  ppmv using our version of Match. Using the PV/Theta approach, the inte-

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grated loss is  $0.4 \pm 0.8$  at 475 K and  $0.8 \pm 0.7$  at 450 K. Again, all three estimates of integrated loss fall within their mutual uncertainties, despite the large differences in ozone loss rates calculated in January.

*Becker et al.* [2000] used a box model to calculate ozone loss rates for the winter/spring of 1991 – 1992. They found that while they are able to reproduce the Match loss rates from mid-February through March, their loss rates for the period at the end of January are significantly smaller, by more than a factor of 2, a result similar to the discrepancy we find in this study between the original Match results and the results from our version of Match. The ozone loss rates of *Becker et al.* [2000] peak at about 4 ppbv/sunlit hour around January 17 with no indication of the large spike in loss rates found in *Rex et al.* [1998] for late January. The ozone loss rates of *Becker et al.* [2000], however, are in quite good agreement with our results for this time period. From their Figure 2, we see that for air parcels descending to 466 K, the ozone mixing ratio changes from about 3.85 ppmv on January 1 to 2.80 ppmv on February 29, 1992, a loss of 1.05 ppmv. Such ozone loss agrees very well with the integrated ozone loss discussed above for 1992.

*Rex et al.* [2003] attempt to explain the large ozone loss rates seen in Match. They use a photochemical box model run along Match trajectories. Assuming total activation of chlorine, they report a maximum loss rate at 475 K in January 1992 of around 5 ppbv/sunlit hour. Such a result, while smaller by a factor of two than the reported ozone loss rates from the original Match technique, are in agreement with the maximum loss rates found in our version of Match.

*Lucic et al.* [1999] use a PV/Theta approach to estimate time-integrated ozone loss at 475 K during the first 20 days of January 1992 when the vortex is well isolated [*Plumb et al.*, 1994]. They found a loss of  $0.32 \pm 0.15$  ppmv, which agrees very well with the ozone loss calculation from both the original Match approach and our version of Match of  $0.3 \pm 0.2$  ppmv. For the same period, our PV/Theta analysis indicates a loss of  $0.5 \pm 0.8$  at 475 K.

*Browell et al.* [1993] report results from their differential absorption lidar (DIAL) study. They observe no polar stratospheric clouds (PSCs) within the polar vortex during the winter of 1991/1992, but do report the development of water ice (type II) PSCs just outside the vortex between Norway and Iceland on January 19, 1992. We note that *Rex et al.* [1998] report their largest ozone loss rates five days later on January 24, 1992. The development of PSCs in this region place them upwind of a number of the European ozonesonde stations included in the Match study, perhaps impacting the results.

Using a combination of their lidar observations and a determination of the total amount of diabatic descent from in situ observations of trace gas species [e.g., *Podolske et al.*, 1993], *Browell et al.* [1993] find a chemical ozone loss of about 23% near 460 K between January and March 1992. This percentage translates to about 0.7 ppmv of ozone, again in reasonable agreement with our integrated Match results.

*Profitt et al.* [1993] use a tracer-tracer correlation between  $N_2O$  and  $O_3$  to deduce ozone loss in the arctic winter vortex for 1991 – 1992. They report their largest ozone loss rates on January 20, 1992 of about 4.2 ppbv/sunlit hour with loss rates of 0.2 – 2.4 ppbv/sunlit hour throughout the rest of the winter season. These loss rates agree reasonably well with the results from our version of Match, but the largest loss rate is more than



a factor of two smaller than that derived from the original Match technique and published by *Rex et al.* [1998] and *von der Gathen* [1995].

*Salawitch et al* [1993] use in situ observations of ClO and BrO from AASE II in conjunction with a photochemical model to determine ozone loss rates. Averaged over the vortex, they find an ozone loss rate in January of 0.4% per day, notably lower than the ozone loss rates they calculate along the ER-2 flight track, which peak at 1.4% per day (about 7.5 ppbv/sunlit hour assuming 6 hours of sunlight, their assumption). They report an integrated ozone loss over the entire winter at 470 K of 0.7 ppmv. Again, these calculated ozone loss rates are consistent with our findings. The large difference between the vortex averaged loss rate and the peak loss rate found by *Salawitch et al.* [1993] suggests a possible explanation for the large ozone loss rates found in *Rex et al.* [1998]: that localized ozone loss rates may briefly yet greatly exceed the rate characteristic of a larger geographic area.

*Braathen et al.* [1994] perform an analysis of ozonesonde data from EASOE and find an average ozone loss rate inside the polar vortex of 0.13 +/- 0.08% per day for air at 475 K during the period January 9 – March 12, 1992. *Rex et al.* [1998] relate that the peak ozone loss rates found using the technique of *Braathen et al.* [1994] yield ozone loss rates of 0.8% per day in mid January, but that such rates are underestimated by 0.1 – 0.35% per day. Correcting for such an underestimate, the peak loss rates become 0.9 – 1.2% per day, in good agreement with the maximum rates reported using ER-2 data *Salawitch et al.* [1993] above.

In summary, a large number of studies and analyses of the ozone losses during the AASE-2/EASOE mission in the winter of 1991 – 1992 converge on roughly the same an-

swers: an integrated ozone loss of 0.7 – 1.2 ppmv between 450 K and 470 K with peak loss rates in mid-January of 4 – 8 ppbv/sunlit hour, with the exception of the original Match results which suggest a peak loss rate of greater than 10 ppbv/sunlit hour. Photochemical models seem to agree well with the observational data and the results from our version of Match.

## 5. A Trajectory Mapping Approach to Match

### 5.1 Methodology

We have noted that employing the various filters in our version of Match effectively eliminates  $\geq 80\%$  of the possible matched sonde pairs and  $>99\%$  of the matched sonde observations. We therefore present an alternate approach to Match that does not rely upon such filters. This approach follows from the development of trajectory mapping as employed by *Morris et al.* [1995, 2000], *Danilin et al.* [2000], and others and was first developed by *Pierce et al.* [1994]. In this approach, all advected air parcels that arrive within the specified Match radius of the new observation are considered matches with the new ozone measurement.

To determine an appropriate vertical scale over which to search for matches, we calculate the autocorrelation of the noise in the ozone profile. Typically, this vertical scale is about 1 km, very similar to the 5 K vertical spacing of the Theta surfaces used in the original Match technique. In our new approach, however, we do not compare a single observation to a single observation. Rather, we use all matches in the cylindrical volume of space around the new observation, about 1 km in height and with a radius of 475 km (1992) or 400 km (2000) (to duplicate the *Rex et al.* [1998, 2002] Match radius criteria). We also permit all parcels initialized in a cluster for each ozonesonde observation to

match in this approach, not just the central parcel. With the large number of qualifying Matches in this alternate approach and with the elimination of the plethora of filters, the statistics for the calculated ozone loss rates are robust. We believe the trajectory mapping approach represents a statistically defensible alternative to the original Match technique.

## 5.2 Results

Figures 12, 13, and 14 show the ozone loss rates as a function of time for the 500 K and 450 K potential temperature surfaces during SOLVE/THESEO 2000 and for the 475 K potential temperature surface during AASE-2/EASEO (1992) respectively. Our data appear in black, blue, and gold (as before) while the *Rex et al.* [2002, 1998] data appear in red. In all three cases, we note much less variability in the average ozone loss rate calculated using the trajectory mapping approach. This result is not surprising: given the substantial increase in the number of Matches through use of this approach over the original Match approach, we expect the ozone loss rates to show a more consistent evolution as the season progresses.

In Figure 12, we see that on the 500 K surface, ozone loss rates begin January near 0, in agreement with the first data point of *Rex et al.* [2002], then slowly increase in magnitude through the middle of February, remaining relatively constant for the rest of the study period. As a result, the trajectory mapping approach appears to produce smaller loss rates in January, but results that are reasonably consistent with those of *Rex et al.* [2002].

Figure 13 shows the loss rates calculated on the 450 K surface for SOLVE/THESEO 2000. On this Theta surface, the loss rates show nearly constant behavior throughout

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January, a transition to a lower loss rate in mid-February, and a slight increase in the loss rate through mid-March. The behavior of the ozone loss rates calculated with the trajectory mapping approach is quite different from the larger changes in ozone loss rates associated with the data from *Rex et al.* [2000].

As a check on the loss rates indicated by the trajectory mapping approach, we again examine the integrated loss over the period of January 20 – March 12. The trajectory mapping approach results in a change of  $-1.2 \pm 0.6$  ppmv of ozone, on the lower end but still well within the range shown in Table 8 of *Newman et al.* [2002].

Finally, Figure 14 shows the loss rates calculated on the 475 K surface for the AASE-2/EASOE mission. This year reveals more variability in the ozone loss rates than seen in 2000. In part, the increased variability may be due to the fact that the launches in 1992 were not coordinated as a part of Match resulting in far fewer coincidences in 1992 compared with 2000 (~1100 versus ~2600). The trajectory mapping results indicate the largest ozone loss rates occur in January 1992 with a steady decrease in the loss thereafter. By the end of February, very little loss is indicated. Such results are quite different in character than the data of *Rex et al.* [1998].

Differences in the results achieved using the trajectory mapping approach and the original Match technique may not be statistically significant if we appropriately take into account and estimate all the sources of error inherent in both approaches. As alluded to in the earlier discussion, evidence from our study suggests that at present, the published uncertainties for the original Match technique may be underestimated substantially and systematically, particularly in January, a month during which very little sunlight is avail-

able and for which the discrepancies with our results and with other studies are the largest.

## 6. Summary and Conclusions

In this study, we have attempted to reproduce the Match studies for the AASE-2/EASOE (1992) and SOLVE/THESEO 2000 (2000) mission periods. We first set out to recreate the Match technique. Although we are unable to reproduce the loss rates published by *Rex et al.* [1998] for January 1992 and have somewhat smaller loss rates in March 2000 than published in *Rex et al.* [2002], the remainder of the data show good agreement with the original results. Furthermore, our sensitivity studies indicate that the actual uncertainties associated with the ozone loss rates from the original Match technique may be much larger than those published, especially during the month of January for which the results are extremely sensitive to the amount of solar illumination and thus to the precise trajectory followed by each air parcel.

To assess the equivalence of our version of the Match technique with the original, we attempt to reproduce Figure 6 of *Rex et al.* [1998], which shows the linear relationship between the ozone change and the amount of sunlight to which the air parcel has been exposed. Our data in Figure 3 indicates excellent agreement with the original data, although our data seem systematically to reveal less ozone loss than the data from *Rex et al.* [1998]. The figure suggests that it may be worthwhile to investigate further the impact of the choice of meteorological fields and corresponding heating rates on the Match results.

Our sensitivity studies indicate that the only Match filter that appear to impact significantly the Match results is that associated with the PV variability along the back tra-

jectory. Filtering out matches that show more than 40% variation in PV along the calculated back trajectory appears to be warranted. Trajectories of up to 14 days can be included in Match analyses with no apparent negative impact on the ozone loss rate calculations. This result is consistent with the methodology of *Schoeberl et al.* [2002], which uses a continuous data injection/trajectory approach with trajectories of up to 90 days duration. In addition, the parcel spreading filter may be unnecessary. Our data indicate that parcels that have spread by up to 3000 km can be included in the Match ozone loss calculations with little negative impact on the results.

In this paper, we apply the PV/Theta approach of *Lait et al.* [2002] to the data from 1992 and find an integrated loss of  $0.8 \pm 0.7$  ppmv at 450 K and of  $0.4 \pm 0.8$  at 475 K over the period January 1 – February 29, 1992. Using our version of Match, we find a loss of  $1.2 \pm 0.4$  ppmv at 475 K as compared to a loss of  $1.2 \pm 0.3$  ppmv at 460 K computed from the original Match technique. Our loss rates show excellent agreement with numerous other papers on AASE-2/EASOE including both in situ observations and photochemical model studies. Discrepancies in ozone loss rates between Match and our PV/Theta calculations lie in part in the fact that the PV/Theta approach only analyzes ozonesondes in the core of the vortex, completely neglecting the ozonesonde observations in the edge region where much of the loss may have occurred in 1992.

When integrating our results for the SOLVE/THESEO 2000 campaign, we find good agreement for the accumulated ozone loss over the January to March period with other studies shown in *Newman et al.* [2002]. Our version of Match yields an integrated ozone loss of  $1.5 \pm 0.6$  ppmv as compared to the loss from the original Match technique of  $1.7 \pm 0.2$  ppmv. While our ozone loss is somewhat smaller in magnitude than

that of *Rex et al.* [2002], it still in the middle of the range of published results (0.7 – 2.3 ppmv).

We suggest an alternative approach to Match based on trajectory mapping. The trajectory mapping approach requires no filtering of the data and relies upon the large number of matches that can be obtained to compensate for the increased uncertainties associated with the individual Matches. Our study indicates that while this approach produces more consistent and slowly varying average ozone loss rates, the loss rates so calculated are smaller in magnitude than those found using the original Match technique. One possible explanation is that Match unintentionally selects highly localized episodes of large ozone losses. The trajectory mapping approach includes far more matches and, therefore, the effect of isolated events of apparently large amounts of ozone loss is mitigated. The largest ozone loss rate of the original Match technique may not be representative of conditions throughout the entire vortex simultaneously, but rather in specific locations in the vortex, as suggested by *Salawitch et al.* [1993] for ER-2 data during AASE-2/EASOE.

In conclusion, we believe that ozone loss rates calculated via the original Match technique for January should be associated with significantly larger uncertainties than the statistical error bars that have been previously published. While the large loss rates found in January may exist somewhere within the polar vortex region, they likely are not representative of conditions throughout the vortex. Furthermore, the large ozone loss rates from Match remain troublingly inconsistent with our current understanding of polar stratospheric chemistry [e.g., *Becker et al.*, 2000], while the smaller ozone loss rates found in our version of Match and in the trajectory mapping approach are more consistent with the

currently accepted polar stratospheric chemistry. Although the Match studies have produced an appealing and consistent picture of high ozone loss rates associated with large areas of cold temperatures (areas that often foster the development of PSCs), the picture may not be so clear when all of the errors are taken into consideration. While Match truly represents a powerful approach to studying ozone loss, it must be applied with great care in order to produce reliable, robust results.

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## Figure Captions

**Figure 1.** The map indicates the 32 stations from which ozonesonde data are included in our version of Match for the analysis of data from SOLVE/THESEO 2000.

**Figure 2.** The change in ozone is plotted as a function of the amount of solar illumination for the 450 K and 500 K potential temperature surfaces combined for the SOLVE/THESEO 2000 period. The data indicate no correlation between the variables, justifying the boot-strap method (see text) for estimating the uncertainty of the ozone loss rate.

**Figure 3.** Our attempt to reproduce Figure 6 from *Rex et al.* [1998] showing the relationship between the average change in ozone and hours of sunlight exposure. The original data from *Rex et al.* [1998] are plotted in red while our data are plotted in blue. Error bars for both data are one standard deviation. Note that we extend the computation beyond the 65 hours of sunlight that appeared in the original figure to ~85 hours. We also show the standard deviation of the amount of solar illumination in each bin using horizontal error bars.

**Figure 4.** The sensitivity of the ozone loss rate on the maximum minus minimum PV difference along the back trajectory (see text) is explored for AASE-2/EASOE (blue) and SOLVE/THESEO 2000 (red). The thick lines represent the mean values while the thin lines represent the means plus and minus one standard deviation.

**Figure 5.** Same as for Figure 4 except that this figure explores the sensitivity of the loss rates to the maximum distance of parcel spreading (see text).

**Figure 6.** Same as for Figure 4 except that this figure explores the sensitivity of the loss rates on the duration of the trajectories in the match (see text).

**Figure 7.** The ozone loss rate as a function of day of the year is shown for the AASE-2/EASEO mission of 1992. The thick colored lines represent loss rates based on calculations of amount of solar illumination that employ different values for the critical solar zenith angle at the day/night boundary. The red squares and associated error bars once again represent the ozone loss rates and uncertainties of *Rex et al.* [1998]. The ozone loss rates appear quite sensitive to this choice in January, but not very sensitive after mid-February.

**Figure 8.** The ozone loss rate as a function of day of the year for the SOLVE/THESEO 2000 mission period of 2000 on the 500 K potential temperature surface. The black dots represent possible loss rates calculated with our version of Match. The thick blue line is the mean of these data while the thin blue lines represent the mean plus and minus one standard deviation of the black dots. The gold curve represents the mean plus and minus one standard deviation computed from the boot-strap technique for a single iteration. The red squares are data from *Rex et al.* [2002] for the same time period. Error bars associ-

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ated with the Rex data are one standard deviation as calculated with the standard statistical approach.

**Figure 9.** An example of how the ozone loss rate is calculated for SOLVE/THESEO 2000 on the 500 K potential temperature surface for the 20-day period of January 12 – February 1, 2000. Each black and red square represents a change in ozone and amount of sunlit time for a single match. The red squares are randomly selected and number half of the matches. The solid black line is the line-of-best fit to the entire data set (both black and red squares). The solid red line is the line-of-best-fit to the red squares only.

**Figure 10.** Same as Figure 8 except for the 450 K potential temperature surface during SOLVE/THESEO 2000.

**Figure 11.** Same as Figure 8 except for the 475 K potential temperature surface during AASE-2/EASOE (1992).

**Figure 12.** As in Figure 8 except using the trajectory mapping approach (see text) for SOLVE/THESEO 2000 on the 500 K potential temperature surface.

**Figure 13.** As in Figure 10 except using the trajectory mapping approach (see text) for SOLVE/THESEO 2000 on the 450 K potential temperature surface.

**Figure 14.** As in Figure 11 except using the trajectory mapping approach (see text) for AASE-2/EASOE on the 475 K potential temperature surface.

Figure 1

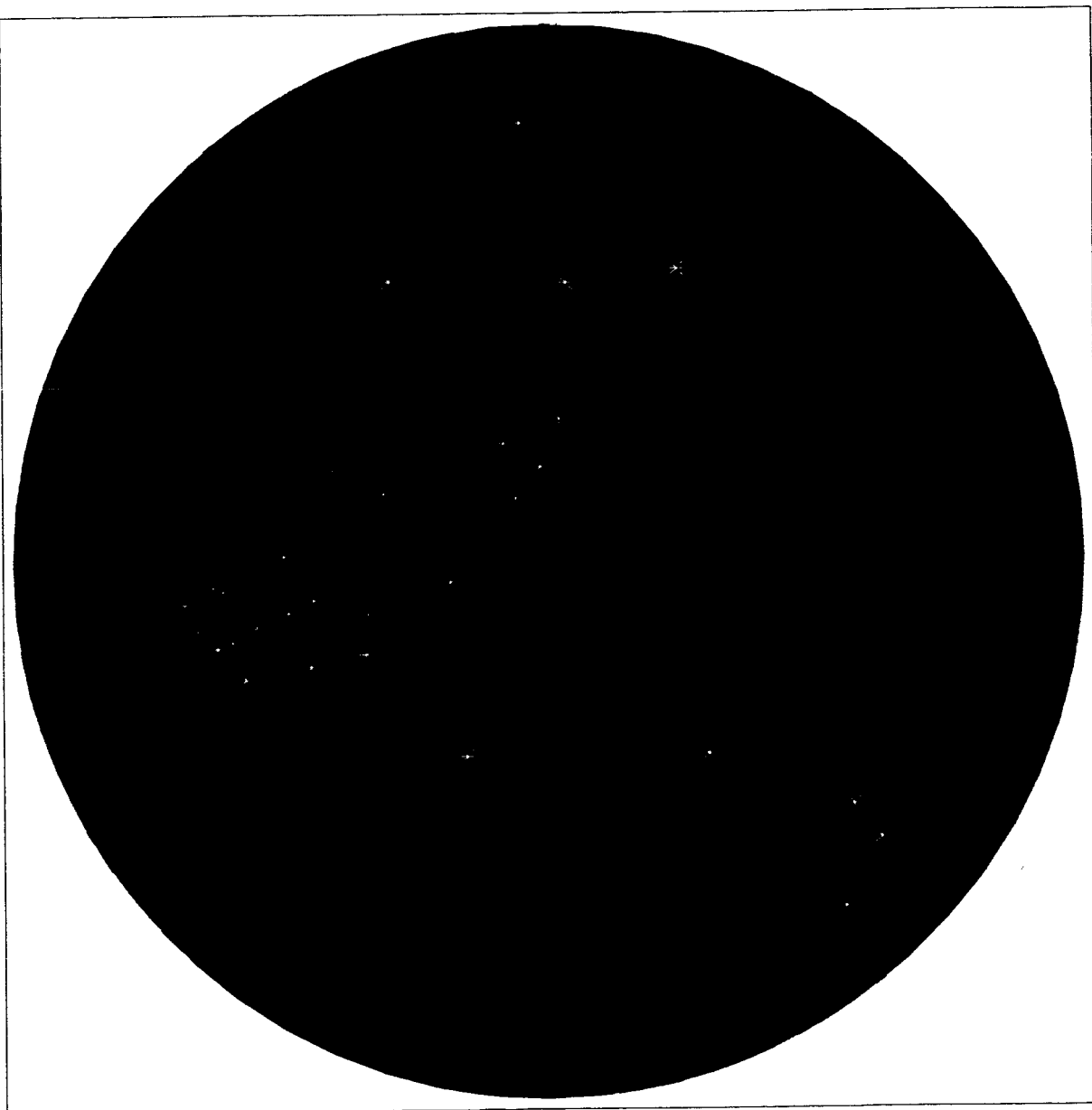


Figure 2

Dependence of  $\Delta O_3$  on Sunlight  
SOLVE at 450K and 500K

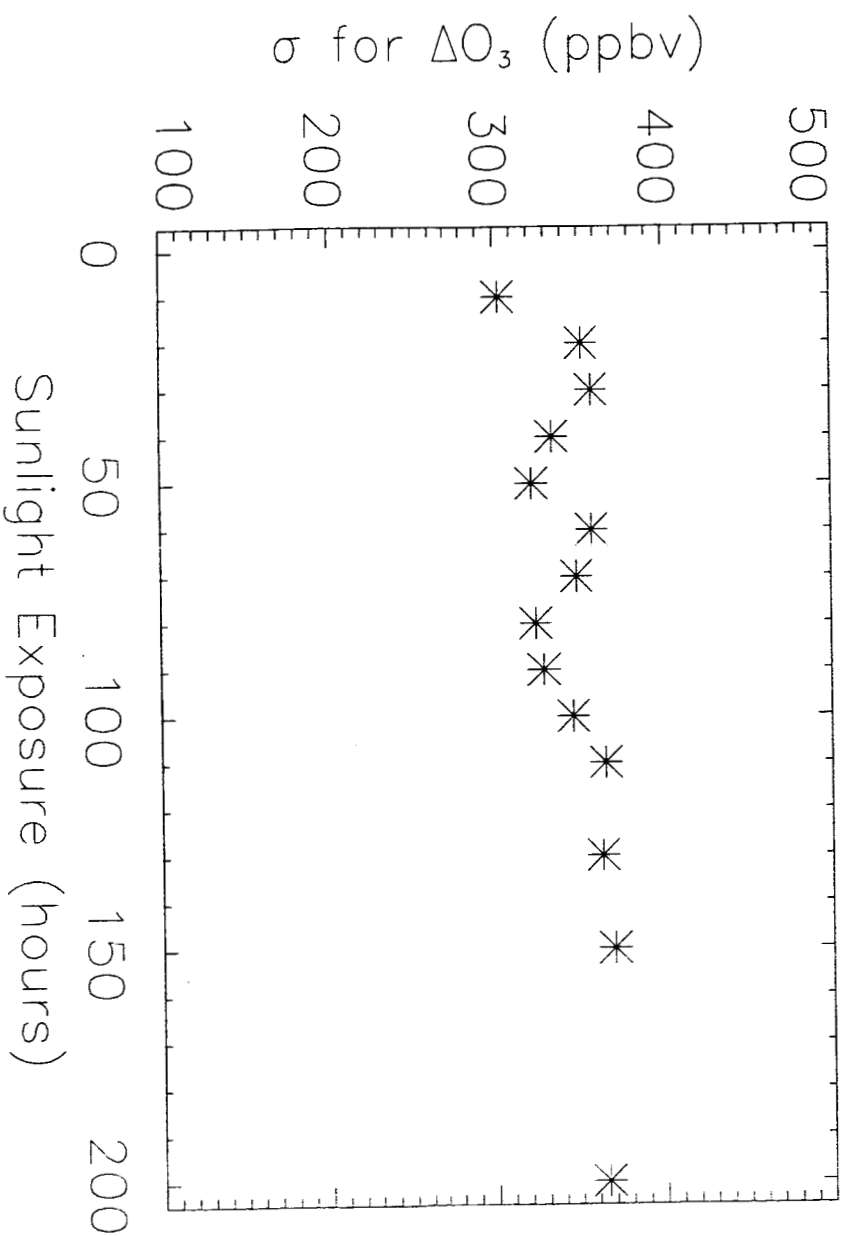




Figure 3

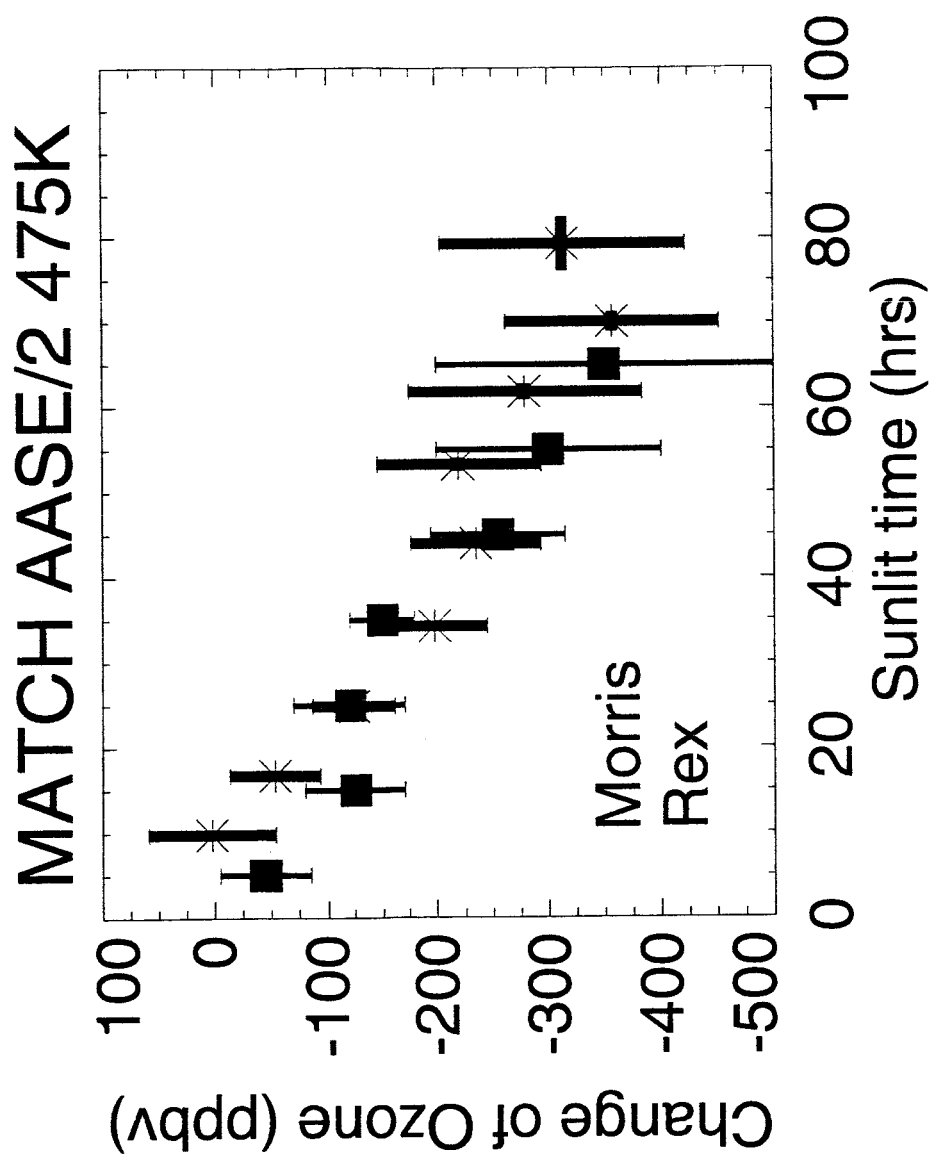


Figure 6 from Rex et al (1998)

Figure 4

# MATCH Sensitivity

## MPV Difference Along Trajectory

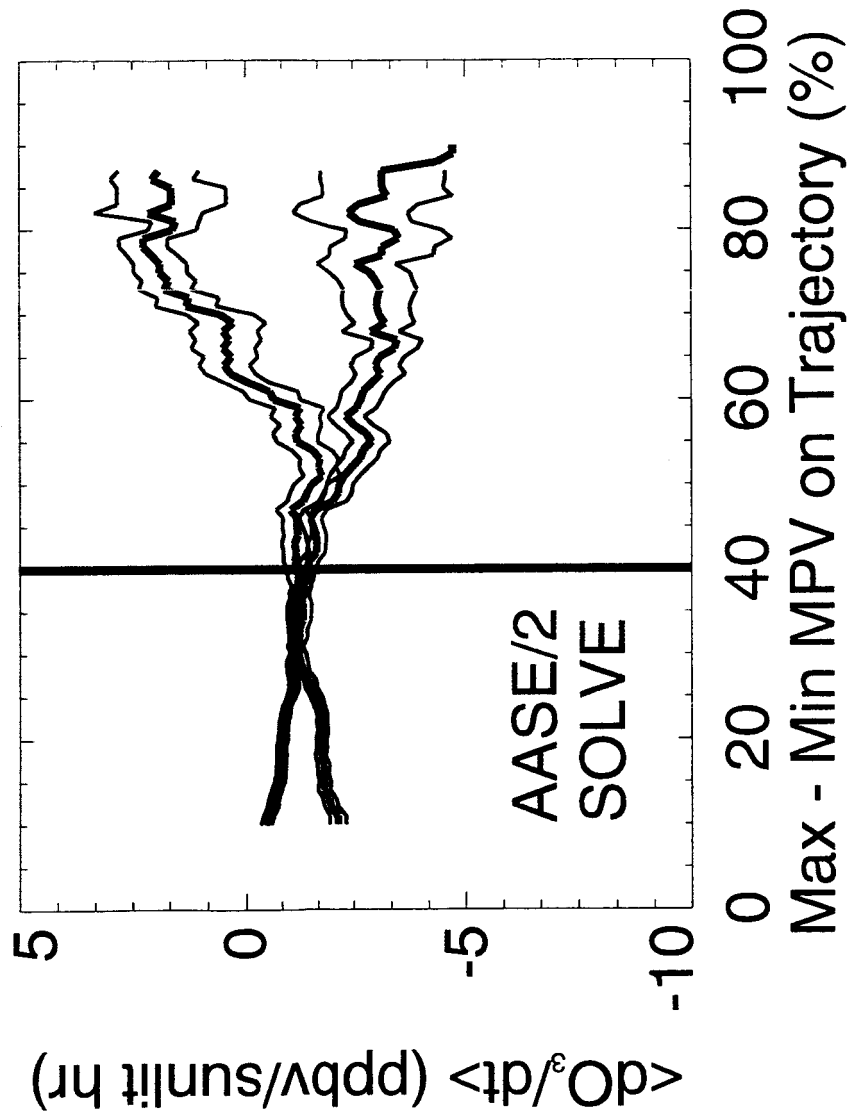


Figure 5

## MATCH Sensitivity

Parcel Spreading Along Trajectory

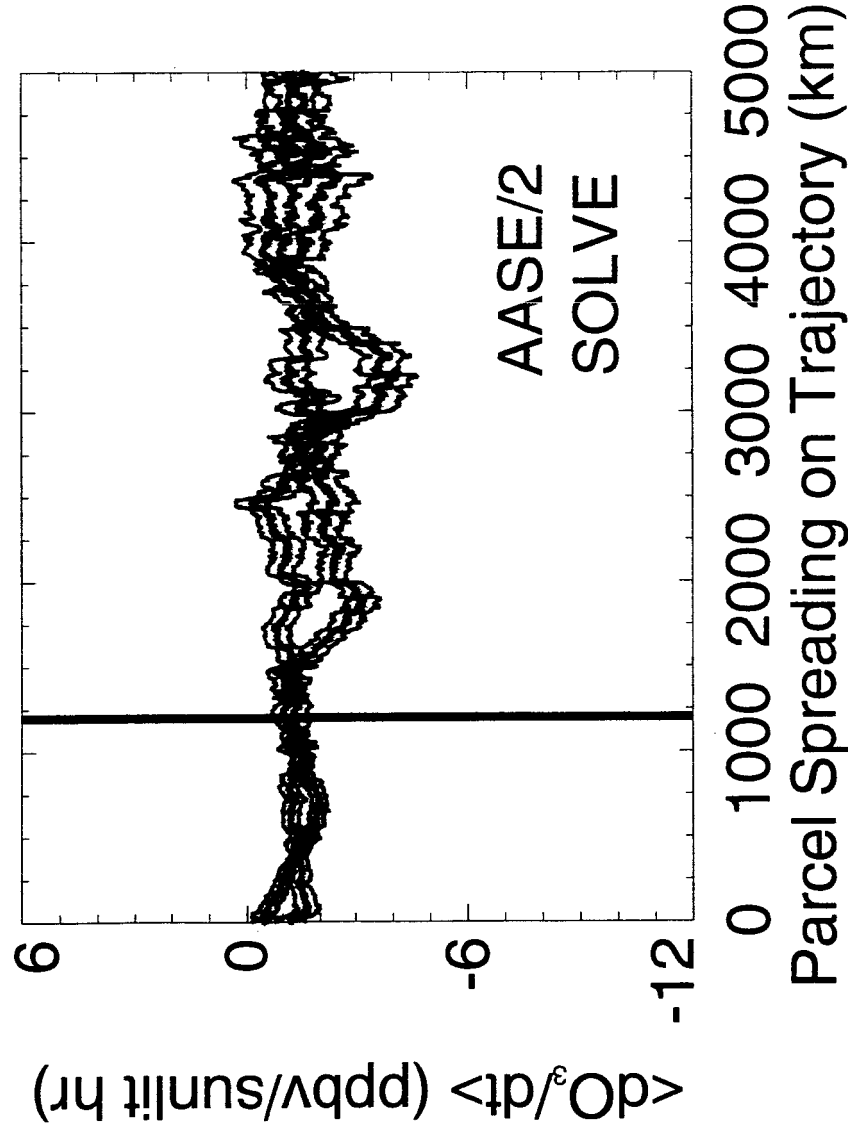


Figure 6

# MATCH Sensitivity

## Trajectory Duration

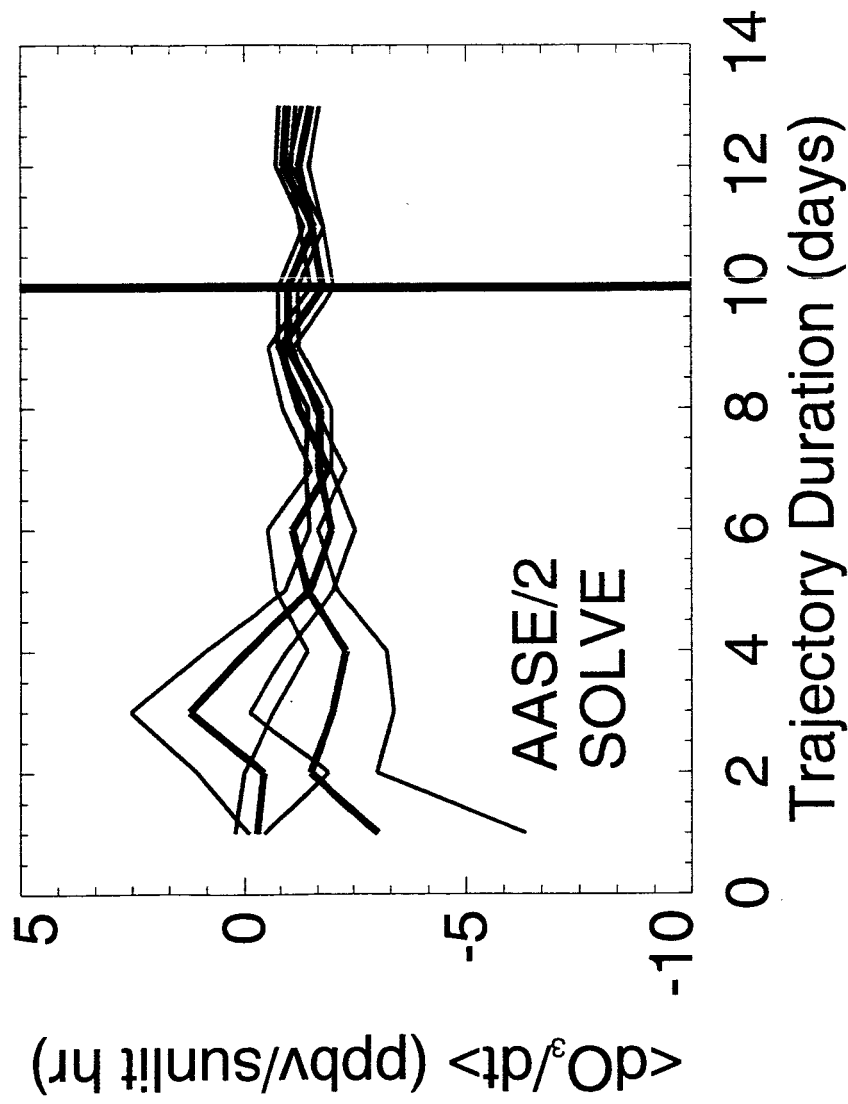


Figure 7

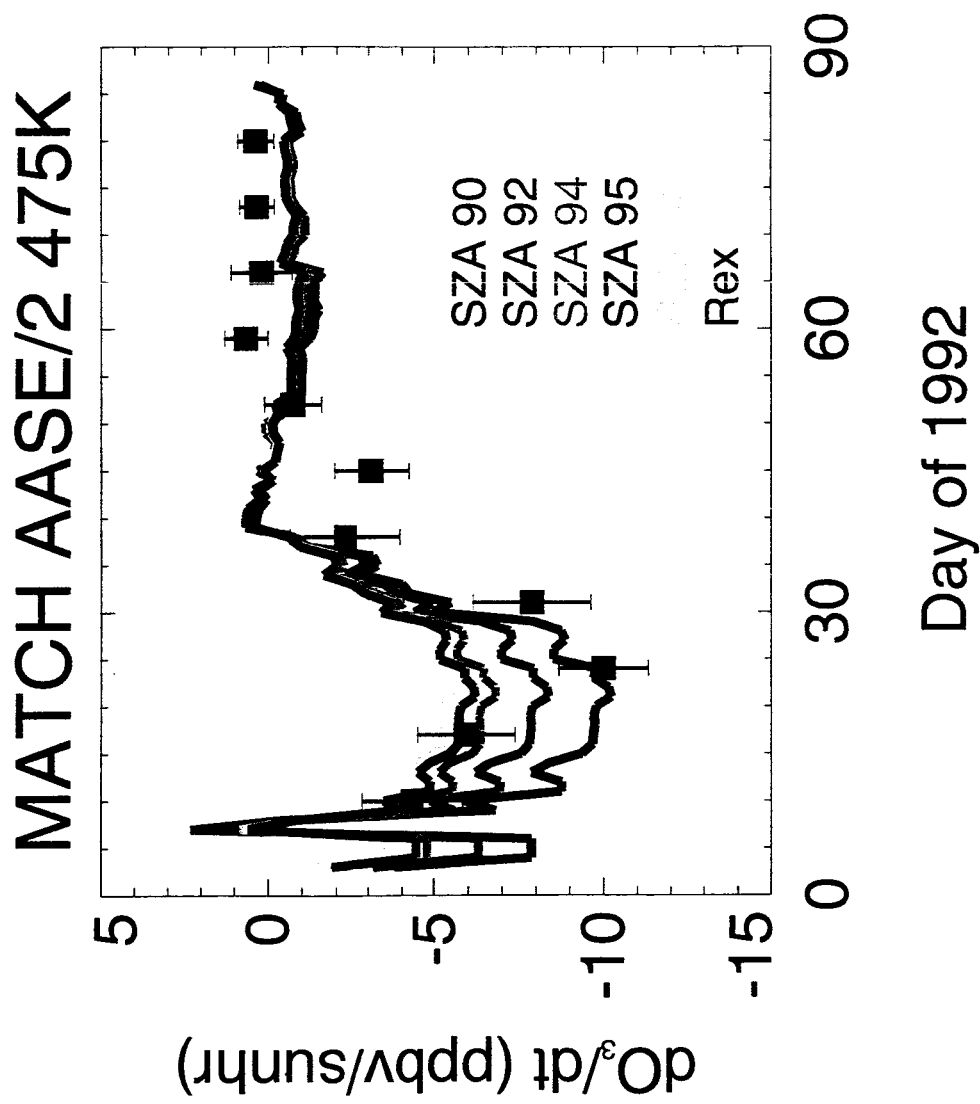


Figure 8

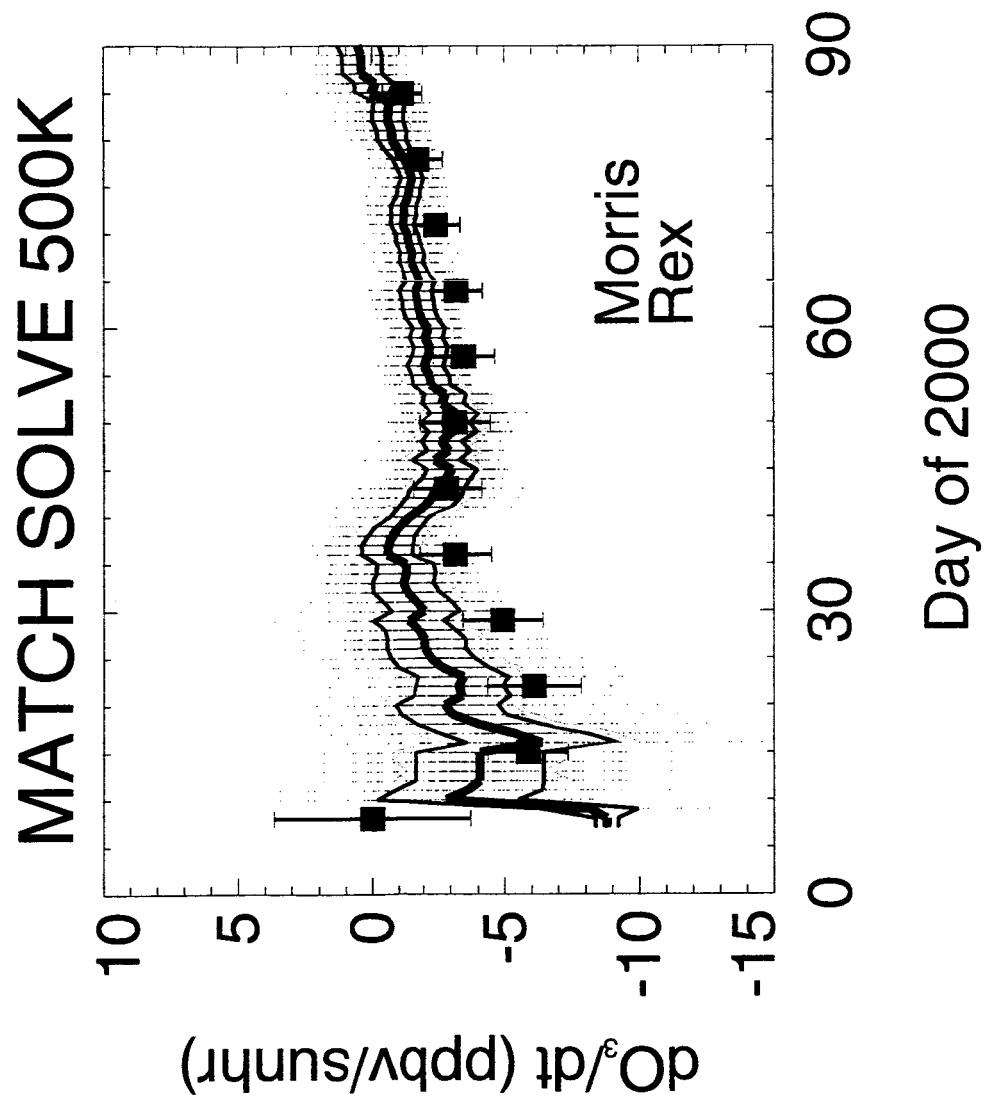
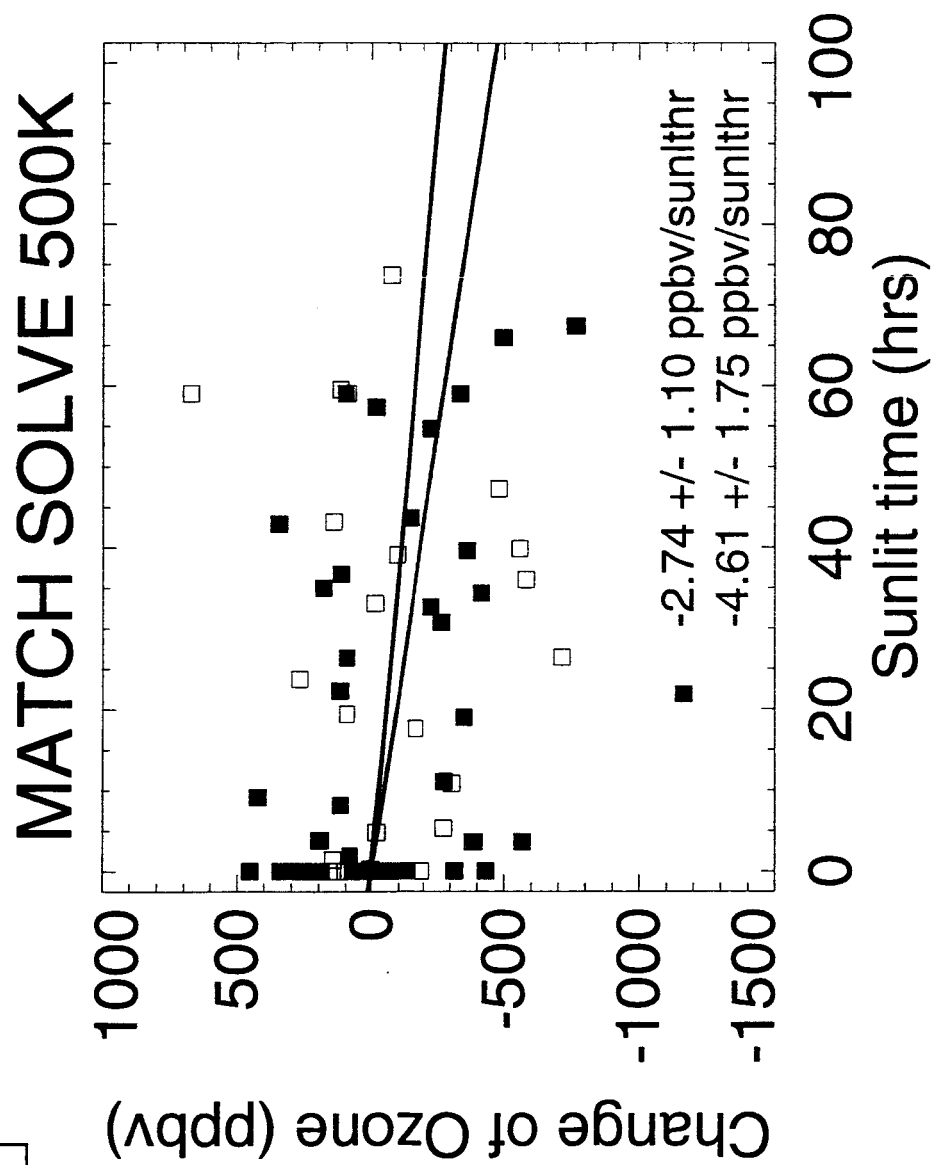


Figure 9



Jan 12 - Feb 1

Figure 10

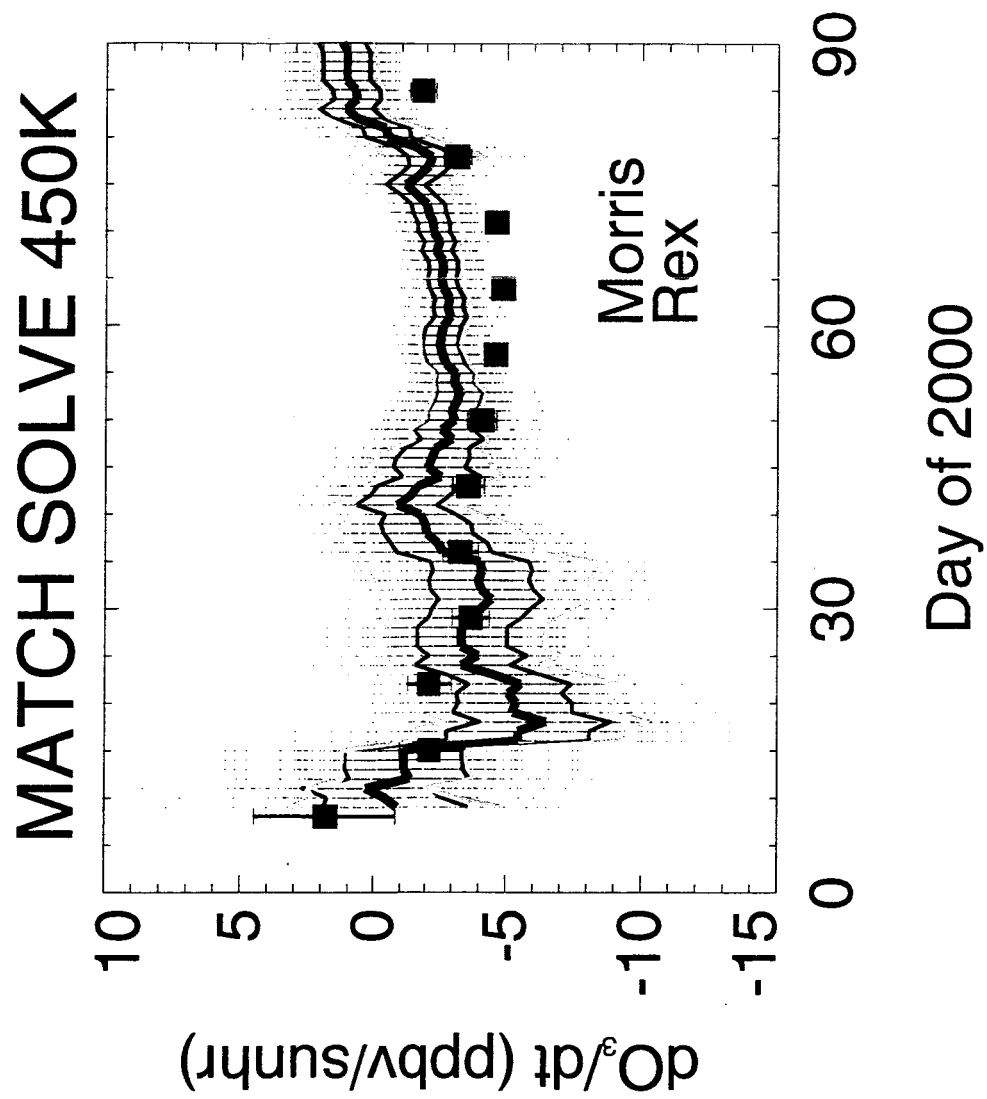




Figure 11

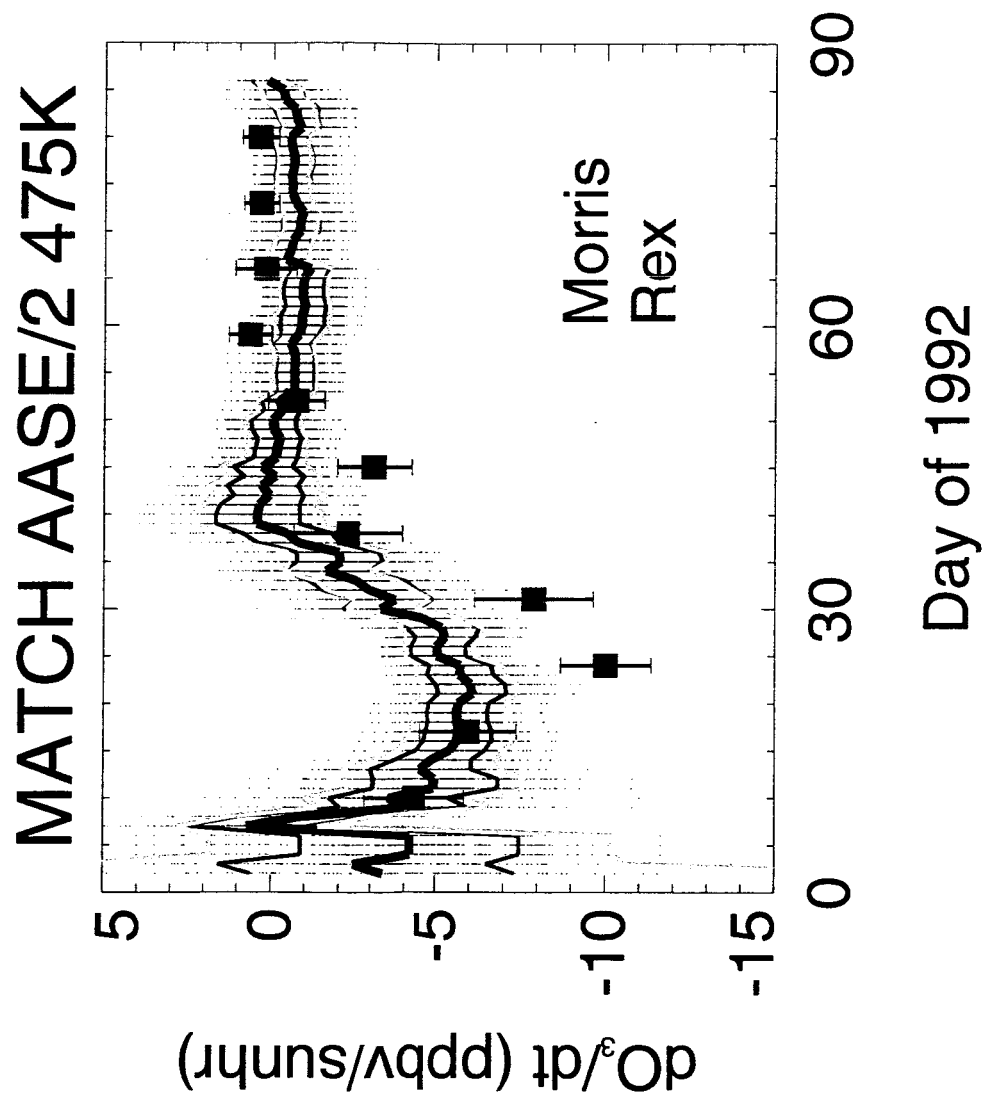


Figure 12

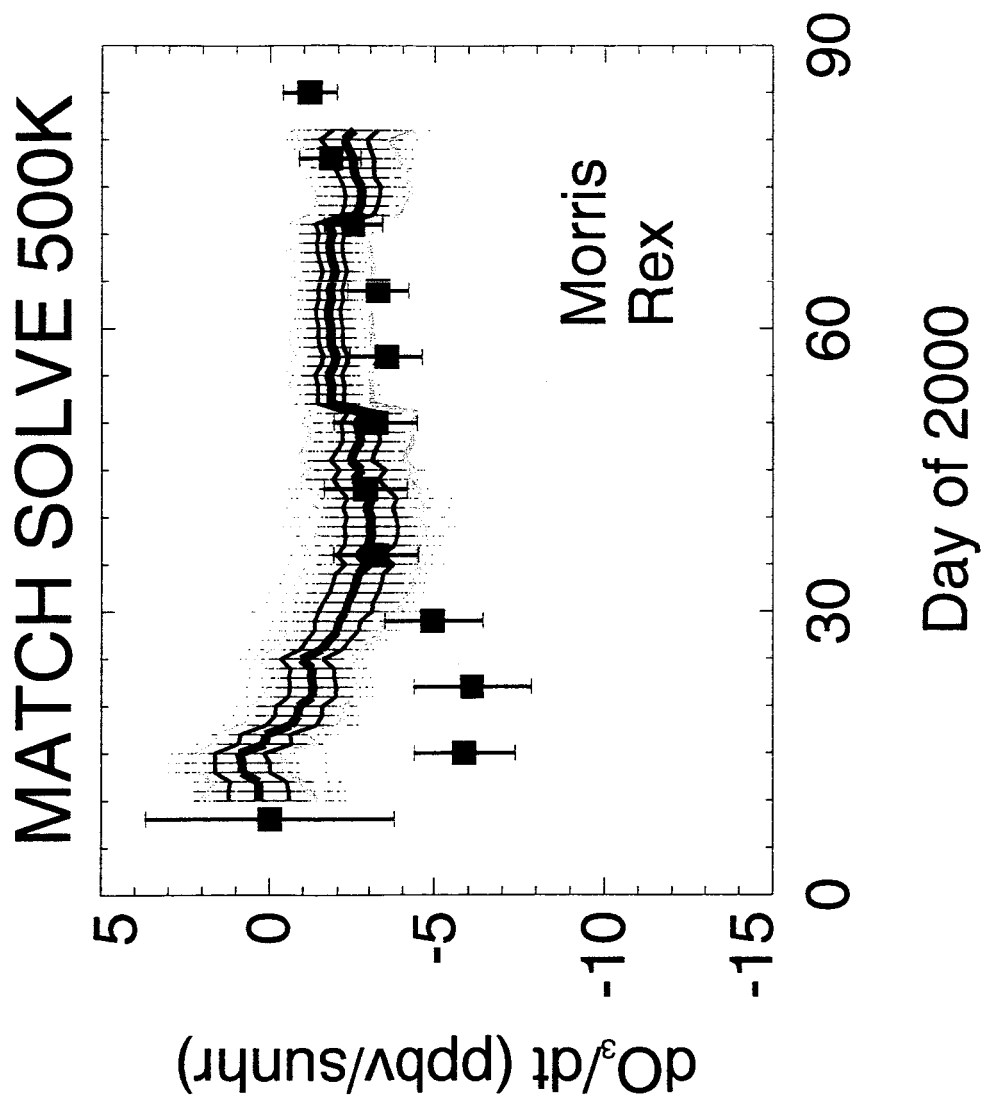


Figure 13

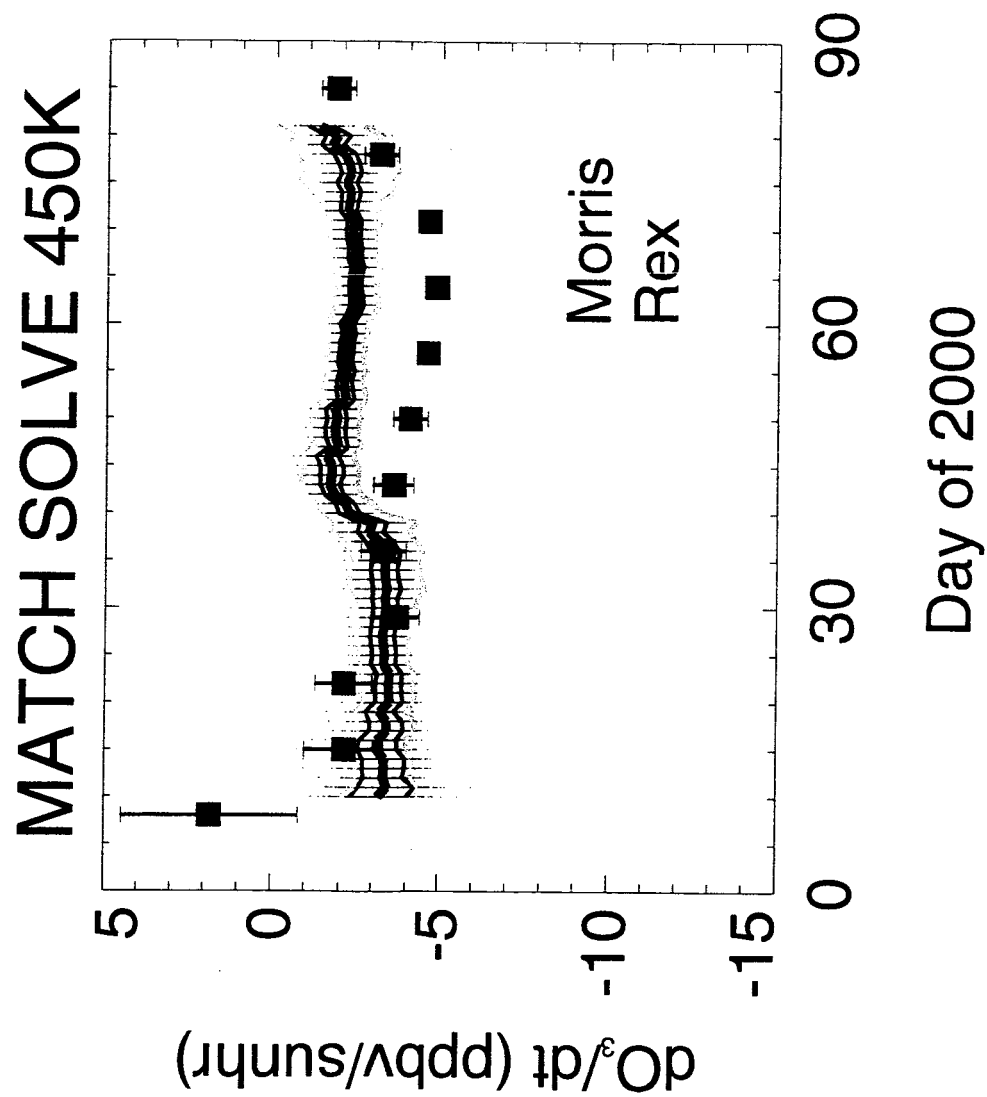


Figure 14

