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MLS CLO OBSERVATIONS AND ARCTIC POLAR VORTEX TEMPERATURES

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Abstract. Analysis of UARS MLS observations in early January 1992 shows a clear relationship between predicted polar stratospheric cloud formation along the back trajectory and elevated ClO amounts. These findings are in good agreement with aircraft observations. The MLS observed variation of ClO amounts within the vortex also fits the pattern of ClO change as a result of air parcel solar exposure and nitric acid photolysis. Outside the polar vortex, the occasional highly elevated ClO appear statistically consistent with MLS measurement noise.

Introduction

The Microwave Limb Sounder (MLS) aboard the Upper Atmosphere Research Satellite (UARS) has been making observations of stratospheric ClO since shortly after the September 12, 1991 launch of UARS. Analysis of these data shows that both the Arctic and Antarctic lower stratospheric polar vortices exhibit elevated amounts of ClO during winter (*Waters et al.*, 1993a,b) consistent with previous aircraft observations (*Anderson et al.*, 1989; *Brune et al.*, 1990).

In addition to the polar observations, however, elevated amounts of ClO are also intermittently reported by MLS at mid-latitudes. These latter measurements could be the result of elevated ClO air transported from the vortex, or local polar stratospheric cloud (PSC) processing which would produce elevated ClO amounts, or simply measurement noise. The purpose of this paper is to investigate the elevated MLS ClO measurements both within and exterior to the vortex using back trajectory analysis. Trajectory analysis provides a powerful tool to validate our understanding of chemical and physical processes within the polar stratosphere (e.g. *Jones et al.* 1992a,b,c). *Schoeberl et al.* [1993a], hereafter S1, found that elevated ClO amounts, observed by aircraft during the three polar aircraft campaigns, occurred on back trajectories with temperatures cold enough to form PSC's. This technique can be used to analyze the MLS measurements, specifically to separate the measurements of ClO which are due to MLS measurement noise and to examine the relation between elevated ClO observations and PSC formation temperatures.

The MLS instrument is described by *Barath et al.,* [1993]. MLS data used here is Version 3, level 3AT which are retrieved on pressure surfaces. The lowest appropriate retrieved pressure surface is at 46 mb for which the MLS ClO measurement noise is estimated to be 0.4 ppbv. To use the trajectory model the data are interpolated to 465 K potential temperature surfaces using NMC temperature analysis.

From December 5, 1991 until January 13, 1992, MLS observed the Arctic polar vortex. PSC formation temperatures were present within the polar vortex during December so that by the beginning of January most of the polar vortex had been "processed" by exposure to PSCs (*Newman et al.*, 1993). By mid-February when UARS yawed back to north pointing, ClO within the vortex was barely elevated (*Waters et al.*, 1993a). PSC temperatures were not observed after late January so the elevated levels of ClO would have subsequently decreased as a result of reaction of ClO with NO_2 which forms with the exposure of HNO_3 to sunlight (S1). Early January 1992 thus appears appropriate for a back trajectory analysis of MLS data.

Since we are interested in examining the relationship between elevated ClO amounts and back trajectory minimum tempeartures, we restrict our analysis to ClO amounts above 0.4 ppbv. From January 1 through the 13, 10 day back trajectories were run for every MLS ClO value greater than 0.4 ppbv north of 40°N on the 465 K potential temperature surface. This amounted to 343 separate trajectories. These data were then statistically analyzed for the correlation with predicted PSC exposure temperatures and total solar exposure, the anal ysis follows that performed by S1 for the aircraft data. The next section describes the results of the back trajectory temperature analysis. The final section describes the relation between ClO measurements and solar exposure.

The early January MLS data

ClO forms the dimer, Cl_2O_2, which is not detected by MLS. Even though small amounts of ClO can still be present at night due to thermal decomposition of the dimer, for all practical purposes, ClO is below the threshold of detectability of MLS at night for individual profiles. Thus MLS ClO values in the lower stratosphere should give a clear picture of the MLS measurement noise.

Figure 1a shows the distribution of all early January data collected between 40°N and 80°N for solar zenith angles greater than 90° (night). This figure clearly shows the precision of the MLS ClO measurement; the Gaussian curve drawn around the histogram of the data has a sigma value of 0.4 ppbv and a mean of 0.06 ppbv. The mean value for the Gaussian distribution used here is based on the daytime aircraft data from AASE II for the region outside the vortex (S1). The slightly higher mean value of the MLS data (0.18 ppbv) may reflect the thermal decomposition of the dimer within the polar vortex, the faint illumination at zenith angles b etween 90° and 94°, or it may be just statistical. The good fit to the Gaussian curve shows that at 465 K the observed precision of the individual values is very close to the estimated precision of 0.4 ppbv.

Figure 1b shows the MLS measurements where the solar zenith angle is less than 90° (day). This figure clearly illustrates the impact of the chemically perturbed stratosphere on the data distribution. There are, in effect, two distributions present: the noise distribution which still fits the Gaussian curve as shown in Figure 1a, and the distribution of enhanced ClO observations most of which lie within the polar vortex. Separating these distributions is discussed below.

Relationship to back trajectory temperatures

For each of the MLS observations where the solar zenith angle was less than 90°, 10 day back trajectories were computed using the trajectory model. To conserve computer resources, the MLS data were grouped into 2.4 hour bins on each observation day and each back trajectory started at the mean GMT starting time for each bin. The temporal size

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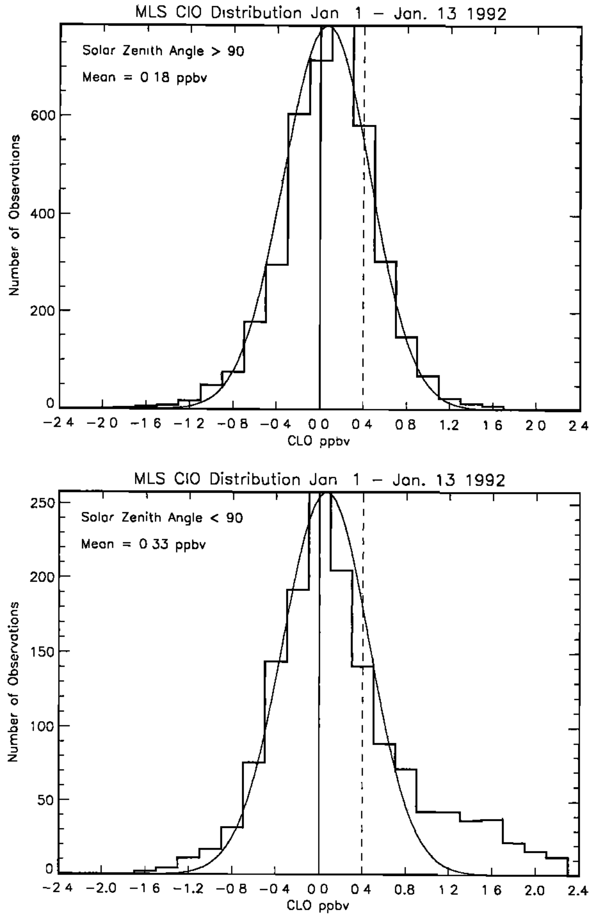

Figure 1. Parts a,b show a histogram of the distribution of Jan 1-13, MLS Level 3at ClO data north of 40° N for solar zenith angles greater and less than 90°, respectively. The smooth curve shows a Gaussian distribution with a 0.4 ppbv standard deviation (the precision of the MLS measurement) centered at 0.06 ppbv which is the approximate aircraft measured mean ClO amount.

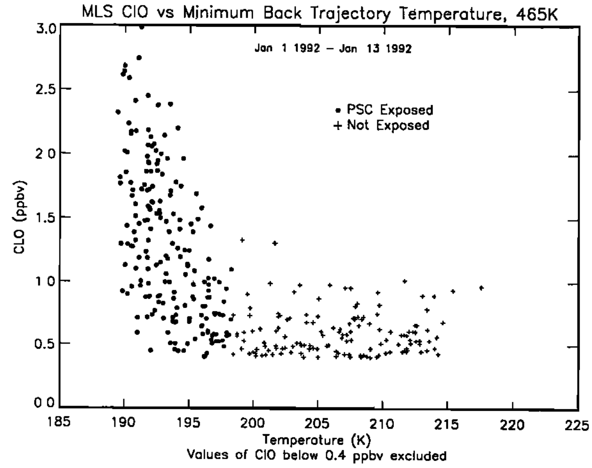

Figure 2. Jan. 1-13 MLS 465 K ClO amounts greater than 0.4 ppbv with solar zenith angles less than 90° plotted against the 10 day back trajectory minimum temperature. Solid circles indicate that the minimum temperature fell below NAT formation temperature assuming 10 ppbv of HNO_3 and 4.5 ppmv of H_2O.

of the data bins was determined by checking the change in the final back trajectory position using exact starting times against the position obtained using the mean bin starting time. For a bin size of 2.4 hours no significant differences in position were found.

For the January 1-13, 1992 period, a total of 343 back trajectories were computed. Figure 2 shows the relationship between elevated ClO amounts and minimum back trajectory temperatures. The solid circles indicate that temperatures at or below the formation temperature of nitric acid trihydrate (NAT) were encountered in the back trajectory. The NAT formation temperature was computed assuming a nitric acid amount of 10 ppbv and a water vapor amount of 4.5 ppbv. These are close to the observed or estimated H_2O and HNO_3 values from the aircraft data (*Kawa et al.*, 1992). Similar to the aircraft results of S1, higher ClO values are observed for air parcels which have probably experienced NAT formation temperatures. However, unlike the aircraft measurements, the lower precision of the MLS instrument produces values above 0.4 ppbv for all observed minimum back trajectory temperatures. The elevated ClO measurements at higher temperatures can be viewed as part of the noise envelope.

Most of the PSC processing takes place within the polar vortex where the stratospheric temperatures are generally coldest. Occasionally, cold temperature regions form at the edge of the polar vortex associated with tropospheric cyclones (stratospheric anticyclones). At 465 K, the polar vortex boundary corresponds approximately to potential vorticity (PV) values of $2.2\times10^{-5} - 2.3\times10^{-5}$ $Km^2/(kg\,s)$. Figure 3 shows the ClO observations plotted against the NMC potential vorticity values. As in Figure 2, the solid circles indicate NAT formation temperatures were encountered. The predominance of "processed" air parcels at high PV values is evident and expected. However, 8% of the parcels outside the vortex with ClO values greater than 0.4 ppbv may also have been exposed to NAT temperatures.

During winter, material is shed from the vortex as it shrinks in size (*McIntyre and Palmer*, 1983; *Butchart and Remsberg 1986*). Also some of the coldest regions of the winter Arctic stratosphere occur near the vortex boundary (*Newman et al.*, 1989). Thus it is not surprising that some of the elevated MLS ClO measurements at mid-latitudes can be traced back to a cold temperature region near the vortex. Is this simply a coincidence, or are such parcels representative of edge vortex processing of air by PSCs, or the shedding of

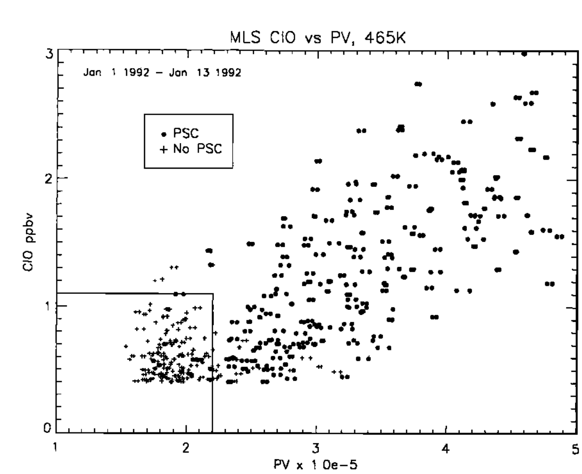

Figure 3. MLS measurements shown in Figure 2 plotted against the potential vorticity at the location of the measurement. PV was computed using NMC balanced winds. Parcels for which the 10 day back trajectory temperature fell below NAT formation temperature are shown as solid circles. PV values below 2.2×10^{-5} $Km^2/(kg\,s)$ K in the northern hemisphere are outside the polar vortex.

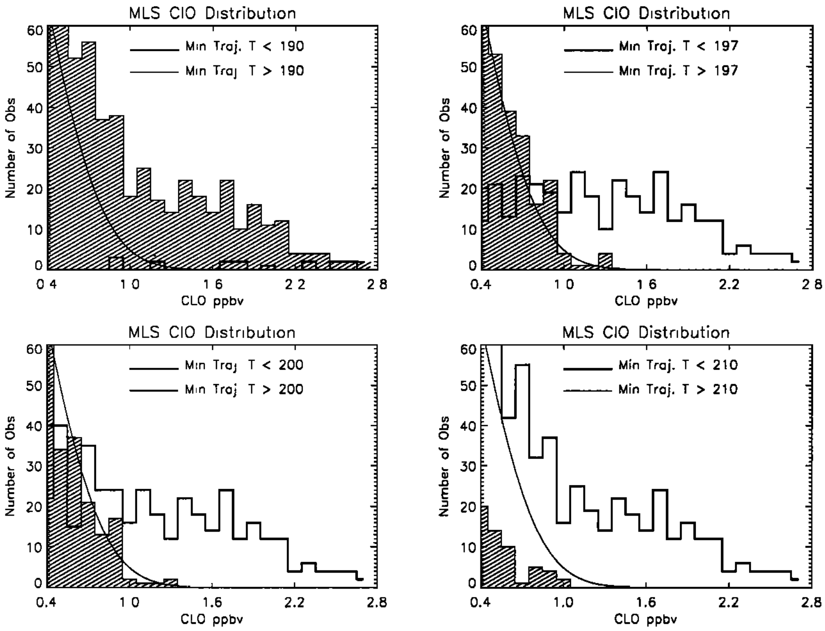

Figure 4. Histograms of MLS ClO measurements from Figure 2 partitioned by minimum back trajectory temperature. Thin curve shows the measurement distribution expected for 0.4 ppbv precision. For the partitioning temperature of 197 K, the measurement noise distribution separates from the perturbed chemical distribution associated with vortex processing consistent with Figure 2.

air by the vortex? In order to answer that question, back trajectories were run for negative MLS ClO values occurring between 40°N and 50°N on January 13. Since negative ClO values are due to MLS noise, it would only be a coincidence that these air parcels encountered NAT formation conditions. About 5% of the air parcels for negative ClO values were found to have been exposed to NAT Thus the 8% extra-vortex "processed" measurements in early January appear to be mostly a result of random chance.

The back trajectory temperatures can be used to separate the noise distribution shown in Figure 1a from the chemically perturbed wing of the distribution in Figure 1b. Figure 4 shows the r.h.s. of Figure 1b with the distribution partitioned by back trajectory minimum temperatures. The 0.4 ppbv 1σ Gaussian curve from Figure 1b is also shown for reference. Four test temperatures are used for partitioning, 190K, 197K, 200K and 210K. The four temperatures are used to illustrate how the distribution shifts with the partitioning temperature. For a partitioning temperature of 197K, the approximate PSC formation temperature, the clearest separation of the noise and PSC processed measurement ensembles appears. The appearance of the secondary distribution is quite consistent with aircraft data (S1) and the results shown in Figure 2. Thus the high ClO measurements seen for back trajectory temperatures greater than about 197 K are consistent with measurement noise. For lower temperatures, the ClO measurement noise is still present, but the mean value of the secondary distribution is greater than 1 ppbv.

Solar exposure

S1 found that the variation of ClO amounts for back trajectories which had experienced NAT formation temperatures could be explained by determining the solar exposure. As mentioned above, once the PSC heterogeneous chemistry produces reactive chlorine and HNO_3, photochemistry will begin to lower ClO. This occurs through the production of NO_2 from HNO_3 and the subsequent formation of $ClONO_2$ by reaction of NO_2 with ClO. The reaction of NO_2 with ClO to form $ClONO_2$ is rapid, and NO_x levels will remain low until most ClO_x has been converted to $ClONO_2$. The amount of NO_x produced from HNO_3 is roughly proportional to the solar exposure as defined in S1.

$$\text{solar exposure} = \int_{t_{PSC}}^{t_{MLS\,Observation}} e^{-\tau/\cos(SZA)}dt \qquad (1)$$

where τ, the optical depth, is set to one, and the ozone overburden is assumed fixed; t_{PSC} is the time the parcel emerges from the PSC and $t_{MLS\,Observation}$ is the MLS measurement time. The integrand is set to zero for solar zenith angles (SZA) greater than 90°. Figure 5 shows the PSC "processed" MLS data points plotted versus solar exposure. Data are

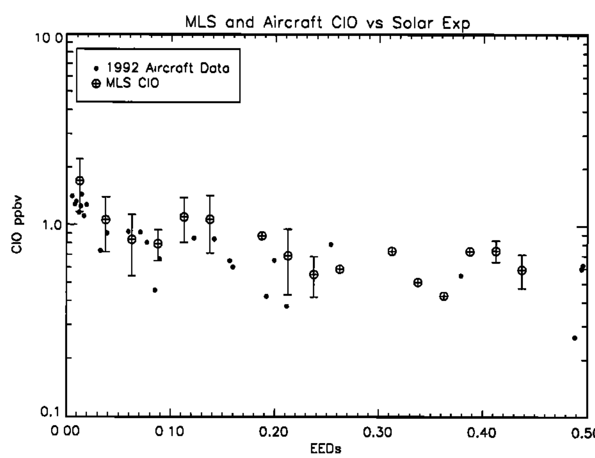

Figure 5. The variation of the MLS ClO measurements with parcel solar exposure shown as open circles with error bars (see text for definition of EED). Only processed air parcels in Figure 2 were considered. MLS data are binned in 0.2 EED intervals. Error vars are the standard deviation of the data within each bin. AASE II aircraft data are shown as points.

averaged and binned in 0.02 EED intervals (an EED is the unity optical depth solar exposure an air parcel receives at the equator at equinox in one day). Error bars indicate the standard deviation. Aircraft data from S1 are also plotted for comparison. The general agreement between the aircraft and MLS data suggests that varying solar exposure is at least partially responsible for the variability in ClO seen in Figures 2 and 4. The scatter in the dependence on solar exposure is partly due to the presence of the dimer not measured in either the aircraft or the MLS observations. Some dimer will always be present in air parcels with elevated ClO. The dependence on solar exposure would thus be clearer if Cl_x (ClO + $2xCl_2O_2$) could be measured. At present this quantity can only be estimated from a model calculation which is beyond the scope of this paper.

Summary

An analysis of the lower stratospheric, northern hemisphere, ClO measurements made by UARS MLS shows that the observations form two distinct distributions. The basic distribution is a noise distribution with 0.4 ppbv σ and near zero mean. This distribution is isolated by examining measurements at night. The second distribution is that formed by the exposure of the air parcels to temperatures cold enough to produce PSCs. This distribution can be separated from the noise distribution by examining the back trajectory minimum temperatures for each observation. The best separation of the two distributions is obtained when the approximate PSC formation temperature (approximately 197 K) is used. Also, nearly all PSC exposed air parcels are found within the polar vortex as defined by the potential vorticity distribution.

Elevated ClO values appear in the MLS data set outside the polar vortex. These isolated values are statistically consistent with MLS instrument noise. Occasionally, back trajectories from these measurements can be traced to the polar vortex; however, it is nearly as likely that negative ClO values (clearly due to measurement noise) can be traced back to the polar vortex as it is that positive ones can. This does not rule out the possibility that an occasional elevated MLS ClO measurement at mid-latitudes might have its origin within the polar vortex, but it appears that a majority of these values whose back trajectories have origins within the vortex have those origins as a result of random chance.

An analysis of the variation of the ClO measurements made by MLS shows a systematic dependence on solar exposure as first noted by S1. Thus MLS supports the notion that variation of ClO within the polar vortex following PSC encounter is due to the reformation of $ClONO_2$ and is proportional to solar exposure.

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