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Comparison of ClO measurements by airborne and spaceborne microwave radiometers in the Arctic winter stratosphere 1993

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Abstract. In February 1993 measurements of chlorine monoxide ClO, one of the key substances in catalytic ozone destruction, were performed over Scandinavia by two microwave receivers, the Submillimeter Atmospheric Sounder (SUMAS) on board the German research aircraft FALCON and the Microwave Limb Sounder (MLS) on board the Upper Atmospheric Research Satellite (UARS). High ClO concentrations (>1 ppb) inside the polar vortex at approximately 20 km altitude were detected by both experiments. A comparison shows good agreement of both sensors in the location of enhanced ClO.

Introduction

Stratospheric ozone is destroyed by reactive chlorine released from CFCs in the process of photo-dissociation. Most stratospheric chlorine generally resides in relatively inert reservoir gases, however inside the polar vortex at low temperatures these reservoirs may break up in heterogeneous chemical reactions taking place on the surface of polar stratospheric clouds (PSC). This process releases chlorine, and under the presence of sunlight a highly effective cycle destroys ozone in the lower stratosphere. Measurements over Antarctica and the Arctic have shown anomalous high concentration of ClO inside the polar vortex (e.g. de Zafra et al. [1989], Anderson et al. [1989], Waters et al. [1993], Crewell et al. [1994]). For a general review see Solomon [1990].

Vertical VMR (Volume-mixing-ratio)-profiles of ClO can be measured by using remote sensing techniques in the millimeter and submillimeter wavelength range. Depending on receiver sensitivity and observing geometry different horizontal and vertical resolution can be achieved. This paper describes results from two different receivers one spaceborne (MLS), and one airborne (SUMAS).

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We will focus on a comparison of ClO in the lower stratosphere, simultaneously measured by MLS and SUMAS on February 15, 1993.

Instrument Description

SUMAS The SUMAS experiment as described in Nett et al. [1991,a,b] and Crewell et al. [1994] measures thermal emission originating from rotational transitions of atmospheric gases at submillimeter wavelengths. Prime target molecules are ozone, HCl and ClO. Due to the high absorption of tropospheric water vapor in the frequency range of interest (e.g. 649.448 GHz for ClO) the sensor needs to be operated on a high flying aircraft at altitudes ≥10 km. The SUMAS has been used successfully in several campaigns, e.g. during the European Arctic Stratospheric Ozone Experiment (EASOE) in the winter 1991/92. The SUMAS observes to the left of the flight direction at an elevation angle of ~12°. Data from the aircraft inertial navigation system are used to keep the observing geometry constant and are stored for later accurate location of the measured profiles. Atmospheric spectra were taken with a cooled Schottky-diode mixer (T_{Sys}(SSB) ≈ 4000K) and an instantaneous bandwidth of 1.2 GHz and analyzed using cascaded filterbanks providing a spectral resolution of 80 MHz in the line wings, 8 channels of 40 MHz and 10 channels of 8 MHz around the line center. In addition an acousto optical spectrometer with 2 MHz resolution over total bandwidth of 600 MHz was available. Sample spectra are shown in the paper by Crewell et al. [1994]. In order to retrieve the ClO-VMR profile the measured spectra have to be corrected for pressure window emission (~15 %) and the influence of other spectral lines, e.g. ozone which contaminates the wings of the ClO-line. The "up-looking" geometry of the SUMAS provides only a limited vertical resolution of 8 km at low altitude and more than 12 km in the upper stratosphere. The horizontal or spatial resolution is mainly given by the integration time needed to obtain a sufficient signal to noise ratio and the aircraft speed. The receiver employed at the time of the campaign allowed the retrieval of a ClO profile every 1000 seconds which translates into a spatial resolution of ~200 km. On recent similar campaigns in February

1994/95 a new super-conducting receiver, developed by the Space Research Organization of the Netherlands (SRON) has been used successfully reducing the measuring time and therefore the spatial resolution to less than 30 km.

Optimal estimation technique [Rodgers, 1976] is used to derive the VMR-profile from the measured spectra. Corresponding to the width of the weighting functions the profiles are given in four independent layers. The a priori profile assumed is typical for the mean global ClO-distribution and does not contain enhanced ClO in the lower stratosphere. The error analysis considers both instrumental noise and uncertainty in spectroscopic parameters. All deconvoluted spectra show a signal-to-noise ratio higher than 10:1. The calibration uncertainty ($< 1\%$) is negligible. We estimate a precision of 0.3 ppbv (2σ range) for the altitude range 20 to 50 km.

MLS The measurement technique of MLS is described by Waters [1993] and the instrument by Barath et al. [1993]. Primary measurements of UARS MLS are ClO, O_3, H_2O and temperature. Secondary measurements include SO_2, HNO_3 and upper tropospheric water vapor. Some initial results from MLS and other UARS experiments are given in a collection of papers in the issue of *Geophys. Res. Lett.* (Vol. 10, Number 12, 1993). Detailed discussion on MLS data processing algorithms and data validation are presented by Waters et al. (Validation of UARS MLS ClO Measurements, submitted to *J. Geophys. Res.*, 1995).

MLS measures ClO by observing thermal emission from the 204.352 GHz ClO line as the instrument field-of-view (FOV) is scanned through the atmospheric limb from above. Atmospheric pressure at the tangent point of the observation path is simultaneously measured by observations of thermal emission from molecular oxygen, and this provides the vertical coordinate on which ClO profiles are retrieved. The FOV vertical extent at the tangent point for the ClO measurements is ~3.5 km, and a complete limb scan is performed every 65 s. The limb scan consists of discrete steps, with step spacing varying between ~1 km in the lower stratosphere to ~5 km in the mesosphere; individual spectra are measured during 1.8 s dwells of the FOV between steps. Horizontal resolution is ~400 km, set by averaging over the atmospheric limb path and orbital movement during the limb scan. The latitudinal coverage is from 34° on one side of the equator to 80° on the other. UARS performs a yaw maneuver at ~36 day intervals, when MLS high latitude coverage switches between north and south. A ClO profile is retrieved from the radiances measured on each limb scan by sequential estimation [Rodgers, 1976]. Vertical variation is represented as piecewise-linear in logarithm of atmospheric pressure, with breakpoints at $10^{(6-n)/3}$ hPa (where n is an integer), resulting in a profile with ~5 km vertical resolution. The a priori profile used for all retrievals is typical of gas-phase model predictions, and thus includes no enhancement of lower stratospheric ClO. An rms uncertainty of 3 ppbv is used for the a priori profile over the full altitude range of 100 to 0.46 hPa. There is no "memory" of the retrieved profile between limb scans, and independent profiles are thus obtained for each limb scan. The ClO measurement precision is estimated for each retrieval, accounting for instrument noise as well as variations and uncertainties in pointing, temperature and other minor effects. Precision of individual measurements over the atmospheric pressure range of 4.6 to 46 hPa is typically 0.5 ppbv rms, and the estimated accuracy is ~15%. Both precision and accuracy become progressively worse outside this range. Waters et al. [1993] give examples of measured ClO spectra, retrieved profiles, and initial results from the arctic winters of 1991–92 and 1992–93. More details of lower stratospheric ClO for the 1992–93 winter as well as results from the 1993–94 winter are presented in Waters et al. [1995]. The MLS ClO results included here were obtained using algorithms which simultaneously retrieve HNO_3 (not doing this can produce lower stratospheric ClO values which are too large by a few tenths ppbv when gas-phase HNO_3 is severely depleted).

Measurement and Results

During February 1993 the polar vortex was well established and cold enough for formation of PSC's. On February 10, 1993, UARS performed a yaw maneuver allowing the MLS to observe the Arctic. On the same day the flight campaign of the SUMAS started. A typical flight of the FALCON takes about 2.5 hours and covers a flight distance of approximately 2,000 km. Figure 1 shows the flight track of the FALCON on February 15. Due to the observing geometry, the observed air parcels are east of the flight track by ~42 km for an altitude of ~20 km. The center points for the MLS tangent points at ~20 km altitude are also shown in Figure 1 together with the potential vorticity (PV) in the 550 K level. From the PV distribution the edge of the polar vortex can be determined at a latitude of ~58° crossing the flight track south of Stockholm.

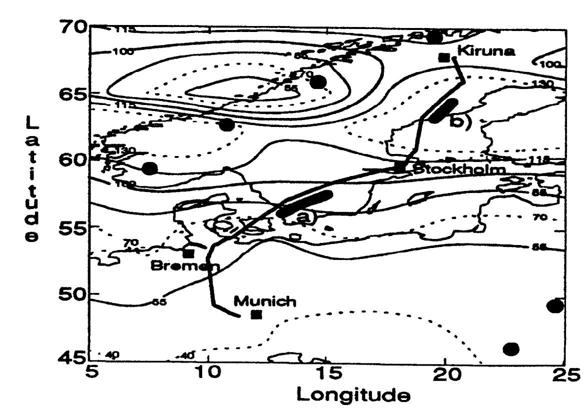

Figure 1. Flight track of the research aircraft FALCON on February 15, 1993 from Kiruna to Munich. Black dots indicate the centers of the MLS tangent points at 20 km altitude. Additionally the potential vorticity [in units of 10-6 Km^2/kgs] at the 550 K level is shown. The (•) indicate the locations of the individual profiles measured by MLS and a) and b) for SUMAS respectively, shown in Figure 4.

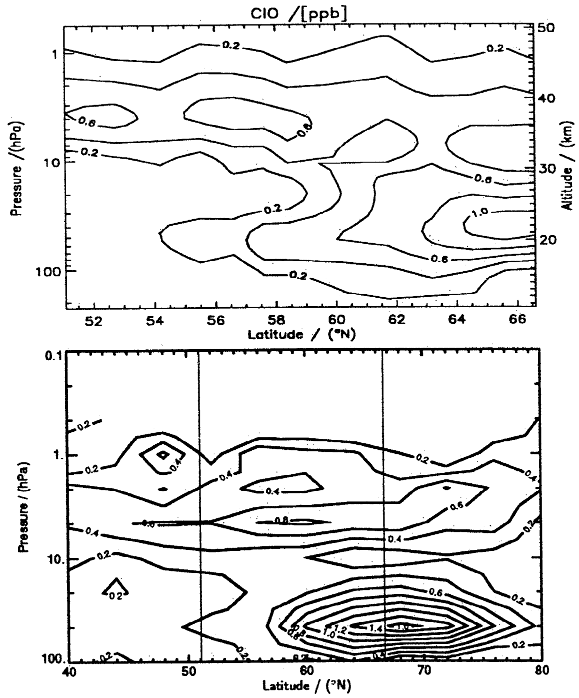

Figure 2. Contour maps for the vertical ClO-distribution in units of ppbv are shown in a) measured on 15 February 1993 by SUMAS and in b) a composite for MLS-data over the 10-15 February 1993 period and the longitude range 0°–15° east. Vertical lines in b) show area displayed in a).

ClO profiles were retrieved for all SUMAS flights, yielding 4–8 profiles per flight. Figure 2a shows the distribution for February 15, 1993 as a function of latitude. The measurements were made over a time period of 10 hours to 15 hours UTC which means close to local noon. For comparison figure 2b shows the distribution for the average of all MLS measurements made between 0 and 45° E longitude during the February 10–15 period of the FALCON campaign. Except for areas with enhanced ClO in the lower stratosphere, averaging the MLS data to reduce noise is necessary to obtain useful comparisons. The MLS measurements were made in daylight up to 76° N and within about 1 hour of local noon up to 74° N. The results from MLS and SUMAS are in good qualitative agreement. The upper stratospheric ClO peak at ~37 km (~4 hPa) is evident, with maximum values of 0.6-0.8 ppbv. Significantly enhanced lower stratospheric ClO is seen northwards of ~58°, with largest values at ~50 hPa (~20 km) height. Figure 3 compares in more detail the SUMAS and MLS results for lower stratospheric ClO on February 15. The SUMAS values retrieved at 20 km altitude, and the MLS values at 46 hPa from the two orbits passing to either side of the FALCON flight track (see Figure 1) are shown. Average values north of ~60° are 1.6 ppbv from MLS and 0.9–1.1 ppbv from SUMAS. Already in Figure 2 and even more so in Figure 3 it is obvious that SUMAS tends to see lower maximum levels of ClO than MLS. This effect is even more clearly seen in Figure 4 where individual profiles outside (Figure 4a) and inside (Figure 4b) the vortex are compared. The gross features are well reproduced by both sensors, however the rather narrow peak of enhanced ClO around 20 km altitude is

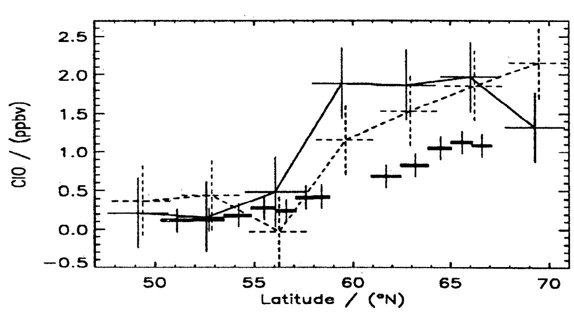

Figure 3. Measurements of low stratospheric ClO over northern Europe on 15 February 1993. The thick line shows values at approximately 20 km height inferred from SUMAS. The crosses show values at approximately 46 hPa pressure (~20 km height) inferred from MLS. The MLS points connected by the solid line are from the measurements track passing over Norway, and the points connected by the dashed line are from the previous orbit passing over eastern Europe and St. Petersburg. The vertical extent of the crosses is the estimated $\pm 1\sigma$ uncertainty, and the horizontal extent is the latitude range over which the measurement is made.

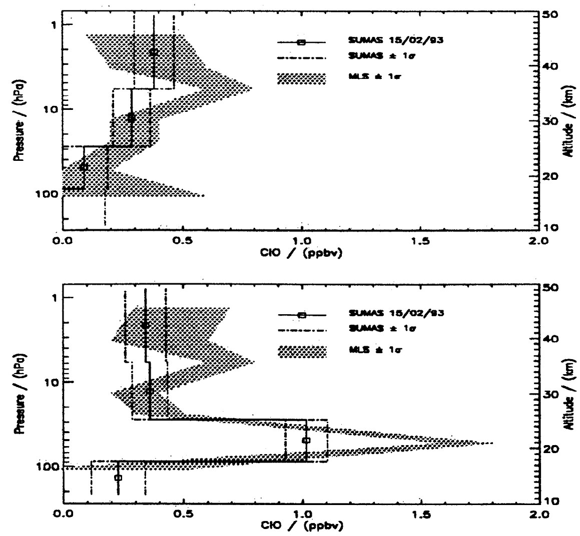

Figure 4. Individual profiles outside a) and inside b) of the vortex are compared. Note the different vertical resolution of MLS (~5 km) and SUMAS (8-12 km).

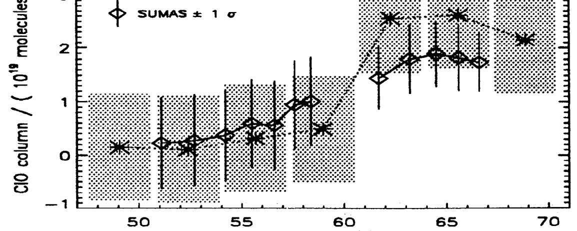

Figure 5. Comparison of ClO column densities derived from MLS- (∗) and SUMAS-data (◇).

smeared out by the more than twice as wide SUMAS weighting functions. This explanation is supported by the result shown in Figure 5 where the column densities are compared. Both sensors agree to within the estimated uncertainties inside and outside of the vortex. The somewhat larger differences at the vortex edge can easily be explained by the structured vortex boundary and the fact that SUMAS and MLS average over considerably different air masses.

Conclusion

In late Arctic winter 1993 two remote sensors operating in different spectral ranges and different observing geometries detected strongly enhanced ClO in the lower stratosphere over Scandinavia. There is good qualitative agreement, and differences in retrieved profiles can be attributed to the different vertical resolutions. The edge of enhanced ClO region is well correlated with the edge of the polar vortex as indicated by steep PV gradients. The temperature field deduced from sonde measurements at several sites along the SUMAS flight track shows a considerable cold region over Scandinavia (70–60° north) with minimum values below 195 K at 20 km which is close to the condensation point of polar stratospheric clouds (PSC's). The ClO concentration maximum detected by both sensors coincides well with this cold region giving strong indication that heterogeneous chemistry on PSC particles has occurred to produce the large ClO abundances in the lower stratosphere.

In this paper major emphasis was placed on the comparison of results from both sensors. However it should be noted that both sensors have their own specific advantages and disadvantages. The spaceborne MLS provides global data with good vertical resolution (<5 km), however the horizontal averaging is rather large as determined by radiative transfer and orbital motion (approximately 400×400 km^2). The airborne SUMAS on the other hand has a much finer horizontal resolution of less than 100 km^2, but a vertical resolution of the order of 10 km. Furthermore the airborne sensor can only be deployed in campaigns over a rather small area. In summary where the MLS has its advantage of providing synoptic type data reliably over long time periods, the SUMAS can very efficiently be used in studying small scale features with very good time resolution over limited observing periods.

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