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Polar stratospheric clouds as deduced from MLS and CLAES measurements

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Abstract. From 30 August 1992 to 3 September 1992 a supersaturated area at 465 K potential temperature (~ 50 hPa) is deduced from MLS water vapour measurements over western Antarctica, where high extinction coefficients measured by CLAES indicate Polar Stratospheric Clouds (PSCs). These PSCs are attributed partly to the effect of an anticyclone located over South America and partly to localized orographic waves, which raise the isentropes and generate rapid adiabatic cooling. A local minimum in column O_3 ($\leq$ 200 DU) is observed in this area, which is believed to be a consequence of the dynamics. Enhanced ClO abundances downstream of the region indicate PSC processing and chlorine activation.

Introduction

The importance of Polar Stratospheric Clouds (PSCs) to the O_3-depleting mechanisms in the Antarctic polar vortex is well known (see *WMO*, 1992). They allow heterogeneous conversion of chlorine from reservoir to reactive forms.

For several days in August and in September 1992 data from the Microwave Limb Sounder (MLS) aboard the Upper Atmosphere Research Satellite (UARS) show an area at 46 hPa in the southern vortex which is supersaturated with respect to ice, i.e. this area is one in which clouds can be anticipated. An examination of aerosols measured by the UARS Cryogenic Limb Array Etalon Spectrometer (CLAES) reveals high extinction coefficients within or in the neighbourhood of this region. In particular the presence of PSCs over western Antarctica is indicated from 30 August 1992 (hereafter denoted by 920830) to 3 September 1992 (920903). We shall show that this episode is related to weather systems in the troposphere and we study its effects upon O_3 and heterogeneous activation of chlorine, in conjunction with UK Meteorological Office (UKMO) geopotential heights, winds, temperature and derived Potential Vorticity (PV).

In general, H_2O measurements at 46 hPa from the MLS 183 GHz radiometer processed with the latest version of the retrieval software (files denoted as "Version 3" in the publicly available data) have an accuracy of ~ 25% and a precision of ~ 0.2 ppmv (*Lahoz et al.*, 1994). However, error estimates associated with retrievals indicate that within the polar vortex in winter the Version 3 data at 46 hPa are dominated by the contribution from climatology. Accordingly, retrievals of H_2O poleward of 50°S have been reprocessed using a non-linear algorithm which can incorporate measurements having high opacity. O_3 and ClO are measured by the MLS 205 GHz radiometer with an accuracy of ~ 20% (*Froidevaux et al.*, 1994) and ~ 15% (*Waters et al.*, 1994), respectively at 46 hPa, and individual profile precisions of ~ 0.2 ppmv for O_3 and ~ 0.4 ppbv for ClO at 46 hPa for Version 3 files. CLAES aerosol measurements are from the UARS processed files (Version 8, not yet publicly available). Extinction coefficients the majority of which are measured with an accuracy of 10-30% at 790 cm^{-1} using blocker 8 (B8) offer the possibility of determining aerosols and PSC events as reported by *Mergenthaler et al.* (1993) while optically thick clouds have larger systematic uncertainties.

MLS and CLAES perform measurements at almost the same locations and local times with a horizontal resolution of ~ 400 km. Profiles are retrieved with vertical resolutions of ~ 5 km and ~ 2.5 km, respectively. Data from MLS and UKMO used in this letter have been linearly interpolated onto the same fixed latitude-longitude grid.

Polar Stratospheric Clouds

We have used the empirical formula from the *Smithsonian Tables* (1958) to estimate e_i, the saturation vapour pressure of water over a plane surface of pure ice. In our analysis, an area is labelled as 'supersaturated' when the MLS H_2O partial pressure, P_{H2O}, exceeds e_i. Figure 1 shows supersaturation (%) (defined as s_i=100×((P_{H2O}/e_i) −1)) and H_2O in excess of supersaturation (ppmv) at 465 K (~ 50 hPa) for the 5-day period: 920830-920903 within a box area defined in Figure 2 over the Palmer Peninsula. Supersaturated

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air is first found on 920830 over a large area (2×10^6 km^2) over the Bellinghausen Sea with a tongue extending over the southern Palmer Peninsula. The supersaturated area then moves eastward over the Palmer Peninsula (920831) to lie east of the Palmer Peninsula on 920901, over the Weddell Sea on 920902. It disappears over the Weddell Sea by 920903.

H_2O exceeds supersaturation by more than 0.5 ppmv (maximum of 1 ppmv) and the associated supersaturation ranges between 20% and 40%. *Toon et al.* (1989) showed theoretically that supersaturation of 20-30% is possible in the stratosphere, dependent upon several parameters such as energy barrier and mean radius of the crystal.

Some points should be borne in mind in the context of the analysis. *i*) The actual temperature fields over Antarctica may be colder than both UKMO and NMC data sets at 50 hPa (*Salter and Merrick*, 1989): a 2-K decrease ($\sim$ 1%) in temperature dramatically increases the estimated supersaturation to 50-70%. *ii*) e_i has been defined with respect to a plane of pure ice; it is evident that the structure of aerosol crystals is far from planar (implying that our estimated supersaturations are overestimates). Also ice present in the stratosphere certainly cannot be considered 'pure', because of the presence of NAT crystals for instance which act as impurities (this would mean that our inferred supersaturation are underestimates, see e.g. *Tabazadeh et al.*, 1994). *iii*) Using a different empirical formula taken from *Marti and Mauersberger* (1993) slightly reduces supersaturation values by 2-3 percentage units. *iv*) Finally, $HNO_3.3H_2O$ or H_2O crystals falling into the lower stratosphere could evaporate within relatively warm layers but thinner than the vertical resolution of MLS (5 km) and this may also alter the calculated supersaturation.

In order to validate the location of our inferred supersaturated areas, we have compared them with the location of measurements of high extinction coefficients by the CLAES instrument, indicative of the presence of PSCs. Extinction coefficients at 46 hPa measured at 790 cm^{-1} are shown in Figure 1. To show the great temperature sensitivity of the spatial extent of the inferred saturated region we have calculated it using both temperature from UKMO (black curve in Figure 1) and UKMO temperature minus 2 K (red curve).

Firstly, we note that the spatial extent of the inferred supersaturated area is much bigger using "T-2 K" than using "T" (T being UKMO temperature). Both areas move eastward and whilst the supersaturated area based on "T" disappears in 920903 that based on "T-2 K" remains over the Weddel Sea. We label extinction coefficients as 'high' when they are greater than 10×10^{-4} km^{-1}. On 920830, high extinction coefficients are almost all located within the "T" and "T-2 K" supersaturated areas. On 920831, the agreement is less impressive maybe because CLAES performed less measurements within the two areas. But high extinction coefficients still appear at the edge of the two supersaturated areas. On 920901, the small number of CLAES measurements prevents any reliable conclusion since there are no measurements within the supersaturated areas. On 920902, the highest extinction coefficients are all located within the "T-2 K" supersaturated area and some of them are even centered within the "T" supersaturated area. On 920903, the agreement is again very good with respect to the "T-2 K" supersaturated area.

Although high CLAES extinction coefficients do not coincide exactly with the supersaturation areas inferred using "T" or "T-2 K", they nevertheless move with the area. "Patchiness" of PSC observations may be coupled to unresolved true "patchiness" in the temperature field. On the other hand, using values of HNO_3 retrieved by MLS (but not yet publicly available) in the formula given by *Fahey et al.* (1989) we calculate that in the neighbourhood of the supersaturated area saturation occurs for typical values of temperature ranging from 184 to 188 K, of H_2O ranging from 2.2 to 3.4 ppmv and of HNO_3 ranging from 1.5 to 4.5 ppbv. The spatial extent of the PSC could therefore be wider than the area strictly defined by supersaturation w.r.t. ice at the analyzed temperature. In conclusion, the extent of the measured supersaturated area w.r.t. ice compared with CLAES aerosol measurements can be resolved by: *i*) a change in the temperature field of about 2 K and/or *ii*) the existence of a HNO_3-saturated area around the supersaturated one.

Formation of the cloud

In general, clouds over western Antarctica may be caused by synoptic scale disturbances in the troposphere (*McKenna et al.*, 1989) and/or orographic waves (*Cariolle et al.*, 1989) that uplift the isentropes and generate rapid adiabatic cooling. UKMO geopotential heights at 500 hPa and temperatures interpolated to 465 K potential temperature ($\sim$ 50 hPa) are shown in Figure 2 for the 5-day period 920830-920903, together with the newly-reprocessed MLS H_2O measurements interpolated to 465 K. The black and red lines represent the supersaturated area when using UKMO temperatures and UKMO temperatures minus 2 K, respectively, while the white one represents the edge of the conservative vortex defined as the PV contour value of -4×10^{-5} Km^{-2}kg^{-1}s^{-1}. The 500-hPa pressure surface is assumed to be representative of the mid-tropospheric weather regimes.

By mid-August 1992, an anticyclone (heights $\geq$ 53 hm) developed over New Zealand (not shown), travelled eastward over South America on 920830 (Figure 2) and over the Atlantic Ocean on 920903. Cyclone-anticyclone pairs are known to play a key role in fluctuations of the tropopause by uplifting (depressing) stratospheric isentropes over anticyclones (cyclones) (see e.g. *McKenna et al.* (1989), *Salby and Callaghan*, 1993). It is particularly obvious that, as the anticyclone pushes below the stratospheric vortex during this 5-day period, isentropes are raised, giving a localised minimum in temperature. Since the center of the vortex is on average dehydrated, the gradient in water vapour is quite strong at the vortex edge. There is some evidence of an intrusion of H_2O-rich air at the vortex edge as can be seen on Figure 2 along the 60°S latitude circle moving eastward from 920830 to 920903 near the location of the northernmost part of our estimated supersaturated area. In addition, at the jet core, MLS H_2O is less than 4.6 ppmv. This is consistent with measurements reported by *Kelly et al.* (1989) in August 1987 over Antarctica. It also supports the claim (*Kelly et al.*, 1990) that southern hemisphere winters in the lower stratosphere are drier than the northern hemisphere winters.

We note that the southernmost area where MLS shows supersaturation and where CLAES aerosols have high extinction coefficients on 920830 and 920831 extends over the southernmost part of the Palmer Peninsula. This is situated quite deeply within the vortex and probably cannot be attributed to the anticyclonic influence, although *Salter and Merrick* (1989) showed an

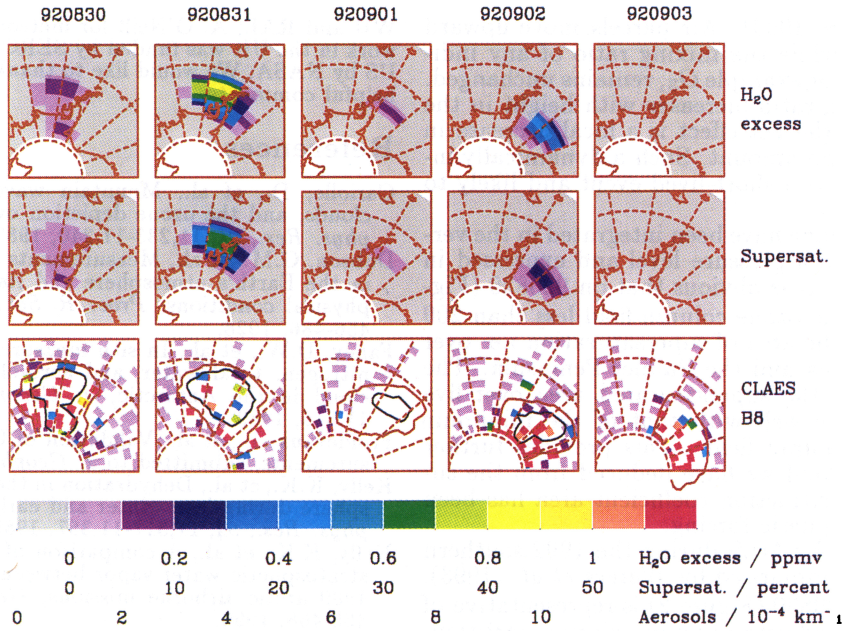

Figure 1. H_2O in excess of supersaturation (ppmv) and supersaturation (%) as deduced from MLS H_2O and UKMO temperature fields from 30 August 1992 to 3 September 1992 at 465 K isentropic temperature (~50 hPa). Aerosol extinction coefficients measured by CLAES using blocker 8 (B8) at 790 cm^{-1} ($\times 10^{-4}$ km^{-1}) during the same period are superimposed over the supersaturated area at 46 hPa using temperature from UKMO (black line) and temperature from UKMO minus 2 K (red line). Polar stereographic projection from 50°S to 79°S where data are plotted only within the black square of the bottom-right map of the figure 2. The Greenwich Meridian coincides with horizontal half-axis at bottom (towards right side).

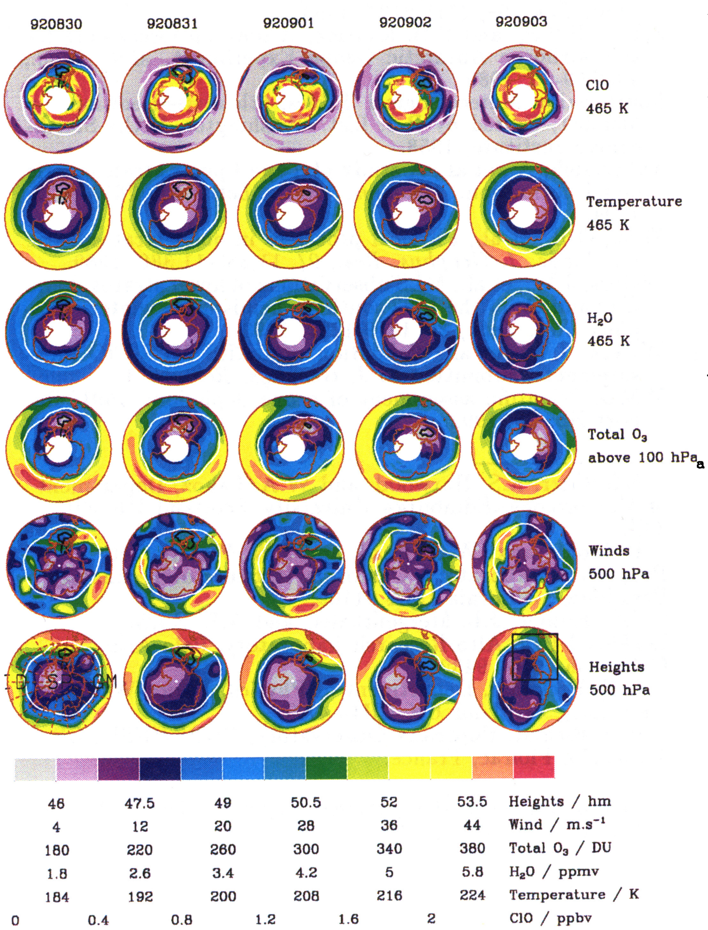

Figure 2. From 30 August 1992 to 3 September 1992, polar stereographic maps from 50°S to the South Pole of: geopotential heights (hm) and winds ($m.s^{-1}$) provided by the UKMO at 500 hPa; MLS column O_3 (DU) integrated upward from the 100-hPa pressure level; MLS H_2O (ppmv) interpolated to 465 K; temperature (K) provided by the UKMO interpolated to 465 K; MLS ClO (ppbv) measurements performed in the descending mode ('daytime' period) interpolated to 465 K. The white line represents the 'edge' of the conservative vortex at 465 K defined by the PV contour value of -4×10^{-5} $K.m^2.Kg^{-1}.s^{-1}$ and the black and red lines represent the supersaturated areas calculated using temperature from UKMO and temperature from UKMO minus 2 K, respectively, except black square in 920903 height map which shows location of figure 1 maps.

example of an anticyclonic ridge near the tropopause which extended all the way to the pole. However, UKMO winds at 500 hPa show a strong jet (winds $\geq$ 28 $m.s^{-1}$) located over the Ellsworth mountains and the elevated Palmer Peninsula. This may have generated mountain waves and produced clouds downstream of the mountains (lee-wave clouds).

Effects upon column O_3 and ClO

The direct effect of the tropospherically-induced adiabatic lifting of the stratospheric isentropes is the lessening of columnar O_3 (e.g. *Dobson et al.*, 1928 and

Salby and Callaghan, 1993). Air parcels move upward to lower pressures while the mixing ratio of any individual constituent, for example O_3, remains unchanged. Since the O_3 mixing ratio increases with height in the lower stratosphere, the net effect is a local decrease in the integrated column amount. Such a dynamically induced O_3 reduction is a short-lived event and likely to be reversible.

Profiles of MLS ozone have been integrated in the vertical from the 100 hPa pressure level and expressed in Dobson Units (DU). It is obvious that on 920830 (Figure 2) the area of the ozone column field less than 200 DU coincides with the area of supersaturation i.e. over the Bellinghausen Sea and the Palmer Peninsula. The eastward motion of the supersaturated area over five days correlates quite well with that of the minimum values in the O_3 column field. This provides further evidence that the cloud we have deduced from the supersaturated, high extinction coefficient area has been generated by tropospheric forcing.

ClO observations by MLS during the 1992 southern winter have been summarised by *Waters et al.* (1993). The ClO field for 920830 (Figure 2) is representative of our current broad understanding of chlorine partitioning. Large ClO abundances within the vortex are generally consistent with chlorine activation by heterogeneous processes on Polar Stratospheric Clouds (PSCs) which form at low temperatures within the vortex. It is probable that the motions responsible for the local maximum of H_2O near the Palmer Peninsula also contribute to the formation of the ClO 'notch' reported by *Waters et al.* (1993) just poleward of this region. The ClO 'notch' can be seen just starting to develop on 920830, and becomes more pronounced later on 920901.

If we focus now on the ClO field located downstream of the cloud from 920830 to 920901, we can see clearly that it shows a local maximum. Since measurements are all made during daytime and have local times which do not significantly vary along a latitude circle, the strong gradient in the ClO field cannot be attributed to diurnal variation. A likely explanation of the ClO increase is that chlorine has been activated by heterogeneous processes taking place within the cloud region giving further evidence of the presence of PSCs.

Conclusions

Measurements from both MLS and CLAES instruments aboard UARS together with UKMO assimilated data sets from 30 August 1992 to 3 September 1992 provide the opportunity of studying: (1) the presence of PSCs at 465 K over western Antarctica, (2) their tropospherically-induced formation, (3) locally-depleted O_3 column fields believed to be a consequence of the dynamics and (4) the chlorine activation by heterogeneous processes through this cloud. Whilst the evidence strongly suggests that at this time and place the clouds we observed are ice-clouds, the spatial relationship between CLAES aerosols and supersaturated areas inferred from MLS/UKMO data shows that either the temperature is colder by 1-2 K than that assimilated by the UKMO or that HNO_3 saturates around the supersaturated area, or a combination of the two.

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