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A 3D SIMULATION OF THE EARLY WINTER DISTRIBUTION OF REACTIVE CHLORINE IN THE NORTH POLAR VORTEX

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Abstract Early in December 1991, high values of ClO are seen by the Microwave Limb Sounder (MLS) on the Upper Atmosphere Research Satellite at latitudes south of areas of temperatures cold enough to form polar stratospheric clouds (PSC's). A three dimensional simulation shows that the heterogeneous conversion of chlorine reservoirs to reactive chlorine on the surfaces of PSC's (processing) takes place at high latitudes. Often the "processed" air must be transported to lower latitudes, where the reactive chlorine is photochemically converted to ClO, to be observed by MLS. In this simulation, one incidence of cold temperatures is associated with an anticyclone, and a second with a cyclone. The transport of processed air associated with the anticyclone is marked by shearing; a decrease in the maximum of the processed air is accompanied by growth of the area influenced by the processing. In contrast, the air processed in the cyclonic event spreads more slowly. This shows that transport and shearing is a crucial element to the evolution of reactive chlorine associated with a processing event. In particular, transport and shearing, as well as photochemical processes, can cause variations in observed ClO.

Introduction

The Microwave Limb Sounder (MLS) on board the Upper Atmosphere Research Satellite (UARS) provides a global view of the appearance and subsequent evolution of high levels of chlorine monoxide within the winter polar vortices. Reactive chlorine species are released from the long-lived reservoir species HCl and $ClONO_2$ through heterogeneous reactions on the surfaces of particles which comprise polar stratospheric clouds (PSC's). Because the heterogeneous reactions are rapid, chlorine is repartitioned (processed) in air which experiences cold temperatures on a relatively small portion of its trajectory.

In early December 1991, MLS observations show sporadic occurrences of high values of ClO [Waters et al., 1993]. During this time, temperatures cold enough for polar stratospheric cloud formation are observed at high latitudes. Although the MLS emission measurement does not require sunlight, ClO concentrations in darkness are generally too small to be detected. The products of heterogeneous reactions are converted to ClO in sunlight, but ClO is small at night due to formation of the dimer Cl_2O_2. Formation of $ClONO_2$ at night is not important because heterogeneous reactions convert almost all NO and NO_2 to HNO_3, limiting the formation of $ClONO_2$.

Three dimensional (3D) model calculations have been used: 1) to determine if the appearance and evolution of high ClO as seen by MLS is consistent with processing by PSC's in the regions of the coldest temperatures; 2) to determine if the behavior of ClO is consistent with a simple model of formation of reactive chlorine in the presence of PSC's and the modeled transport, or if chemical changes such as the reformation of reservoir $ClONO_2$ are suggested by the ClO observations [Waters et al., 1993]

Model

The NASA/Goddard three dimensional chemistry and transport model uses winds and temperatures from the STRATAN stratospheric data assimilation system for transport, and thus model results may be sensibly compared with measurements [Rood et al., 1989; 1991; 1992]. The horizontal resolution of the model is 2° lat by 2.5° long; the vertical spacing is about 3.5 km in the stratosphere. The transport schemes are discussed by Allen et al. [1991]. In previous applications, good agreement between satellite measurements and modeled ozone [e.g., Rood et al., 1991] indicates that the model represents the middle and high latitude synoptic and planetary scale features which are important to transport in the lower stratosphere.

Calculations

These experiments are similar to those performed for the winter of 1989 [Douglass et al.,1991; Kaye et al., 1991]. HCl is initialized by mapping profiles from the GSFC two dimensional (2D) model onto the 3D field using potential temperature as the vertical coordinate and potential vorticity to characterize polar, middle latitude and tropical air [Douglass et al., 1990]. The HCl evolution is calculated with the photochemical production and loss approximated by zonally averaged values from the 2D model, updated every two weeks. A second simulation is performed, using production and loss from the 2D model, and including an additional loss with a 0.5 day time scale whenever the temperature falls below a critical level. This time scale is based on the observation that the appearance of areas of high ClO closely follows the observation of temperatures cold enough for PSC formation. The critical temperatures T_c used here are 201, 199, 197, 194 K at 175, 91, 53, 31 hPa, about 2K warmer than the temperatures for Type I PSCs taken from laboratory experiments of Hanson and Mauersberger [1988]. The warmer values compensate for a

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slight warm bias in the assimilation temperature fields. The difference between these two simulations (DELTA given in ppbv) is a measure of the effect of heterogeneous reactions on HCl. Calculations with smaller values for T_c show fewer areas of processing and smaller values of DELTA. For fixed values of T_c, the magnitude of DELTA increases (decreases) as the time scale for heterogeneous processing is decreased (increased).

Large values of DELTA, when in sunlight, are expected to be associated with large values of ClO. The relationship is qualitative for several reasons. First, the primary reaction which releases chlorine from the reservoir species involves HCl and $ClONO_2$; these experiments only show the HCl changes. Second, the amount of HCl processed depends directly on the HCl initial condition. For these calculations, we presume formation of $ClONO_2$ is negligible. The formation of HCl for high levels of reactive chlorine has been evaluated using a photochemical box model. This effect is small at high latitude during winter and is not considered in the following discussion.

Results

Figure 1 shows ClO synoptic maps generated from MLS measurements at 46 hPa for Dec. 7-15 1991. These data were produced using MLS software version 4.11 and the asynoptic mapping procedure described by Elson et al. [1993]. Contours are shown for ClO values larger than 0.7 ppbv in order to be above the approximate detection limit for individual measurements. These ClO maps are daytime values at the same local time for each longitude. These maps show no diurnal variation with longitude at a particular latitude, as further discussed by Elson et al. [1993].

On Dec. 7, the largest ClO values are seen at about 60°N, 45°E. On Dec. 8, a somewhat larger and more intense ClO maximum is seen at about the same location. On Dec. 9, this feature has decreased in magnitude, and is located

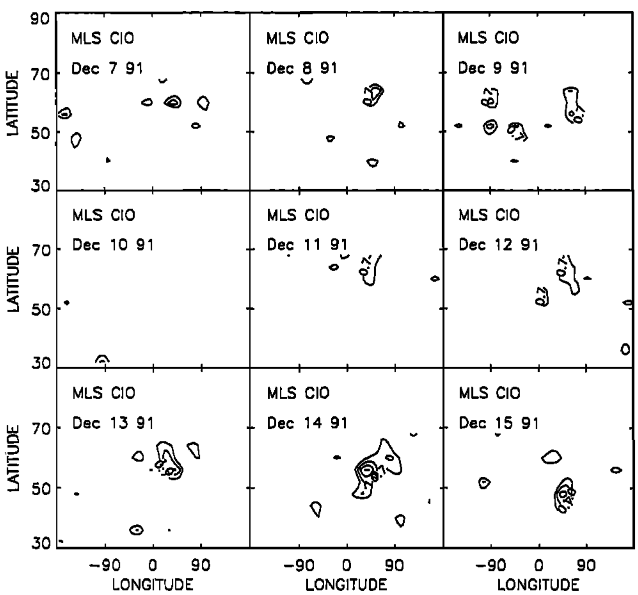

Fig. 1. MLS observations of enhanced ClO (ppbv) in the northern hemisphere at 46 hPa for Dec 7-15, 1991; contours 0.7 + 0.1N ppbv

further eastward. There are also high values between 45°N and 70°N at 45° and 90°W. There are no significant features on Dec. 10. On Dec. 11, a signal appears between 55° and 65°N and 0° and 60°E. The area increases somewhat on Dec. 12. On Dec. 13 there is again a large ClO signal which appears to intensify on Dec. 14. The signal has decreased substantially by Dec. 15.

These ClO measurements may be compared with modeled DELTA during the same time period given in Figure 2. Also given on this figure are the locations of MLS observations for solar zenith angle less than 90° which take place throughout the day. The DELTA contours are the result of all processing which takes place up to 00 GMT on the day of the plot, and are not necessarily collocated with the bold contour which indicates the area where $T \leq T_c$ at 00 GMT on that day. There is not a one to one correspondence between observations of enhanced ClO and areas where the model indicates that processed air could be observed. Here we focus on the observations which show

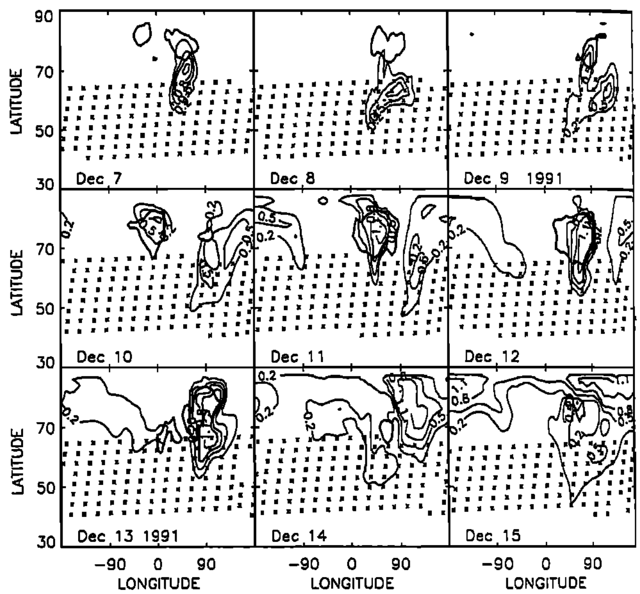

Fig. 2. Model contours of DELTA (ppbv) at 50 hPa, same period as the MLS observations given in fig. 1. The x's indicate the locations of sunlit measurements. The bold contour is the region where the temperature $T < T_c$ (criterion for heterogeneous processing).

the largest area of enhanced ClO, and use the model to infer their relationship to areas of cold temperatures and to each other.

The model shows that the high ClO values are associated with separate processing events which for the most part take place north of the illuminated latitudes viewed by MLS. The high ClO on Dec. 7-8 is seen when the maximum of the processing event, which began in the model on Dec. 6, reaches 55°-65°N latitude where it is sampled by MLS. On Dec. 9, the data show that the ClO maximum has decreased and is located slightly to the east. The model on Dec. 9 indicates that the DELTA maximum has been transported towards the east and north. On Dec. 10, the DELTA maximum in the model is reduced, and the large area enclosed by the 0.5 contour has been transported north and east, leaving a region of smaller values to be sampled by MLS which shows no large areas with ClO of 0.7 ppbv. This could be due to chemical processes which decrease ClO or more rapid northward and eastward transport than produced by the model. The high ClO features in the western hemisphere between 50° and 70° N on Dec. 9 persist for only 1 day and are apparently not associated with regions of PSC temperatures in the STRATAN analysis.

The second processing event is apparent in the model Dec. 10. The processed air is transported southward and the area with $T \leq T_c$ grows until on Dec. 11-12 the area with high DELTA has reached a sunlit latitude sampled by MLS. On Dec. 13 this second area of processed air has begun to move east and north, and a new area of cold temperatures has appeared. The high ClO values on Dec. 13-14 are apparently associated with this new processing event which is accompanied by rapid southward motion as seen by the modeled DELTA on Dec. 14. This event is much more intense in the data than in the model, which suggests that either the duration of the temperatures with $T < T_c$ is too short or the time constant for the heterogeneous loss of HCl is too long. This processed air has also moved northward by Dec. 15, which accounts for the decrease in the area and magnitude of the ClO signal seen by MLS.

The appearance and disappearance of high values of ClO at 55°-65°N is consistent with the model transport. The importance of transport to observations is also inferred from the subsequent behavior of the Dec. 6 and Dec. 10 processing events. The contour of the maximum value of DELTA is 1.1 for both events, and for both events the area where $T \leq T_c$ lasts several days. In the first event the 1.1 contour is present only on Dec. 7; transport processes reduce the maximum so that by Dec. 12 there is a large area enclosed by the 0.2 contour, a small area enclosed by the 0.5 contour, and no higher contours. In the second event, there is evidently less shearing as the area of the maximum grows and is maintained for a much longer period. This is substantiated by comparison of the appearance of the processed air from the two events five days after they appear in the model. The first event has been spread over a much broader area. The maxima associated with the two events for several successive days and the ratio of the maximum for each day to the maximum for the event are given in Table 1. Assuming that these two events take place in a similar chemical environment, the ClO maxima from the first event are clearly reduced by more rapid mixing than the ClO maxima produced by the second event.

TABLE 1. The maximum for each day associated with each event, and the ratio of the daily maxima to the maximum for each event

Event 1			Event 2		
Day	Max	Ratio	Day	Max	Ratio
6	0.9	0.75	11	1.2	0.86
7	1.2	1	12	1.4	1
8	1.0	0.83	13	1.4	1
9	0.9	0.75	14	1.4	1
10	0.8	0.67	15	1.3	0.93

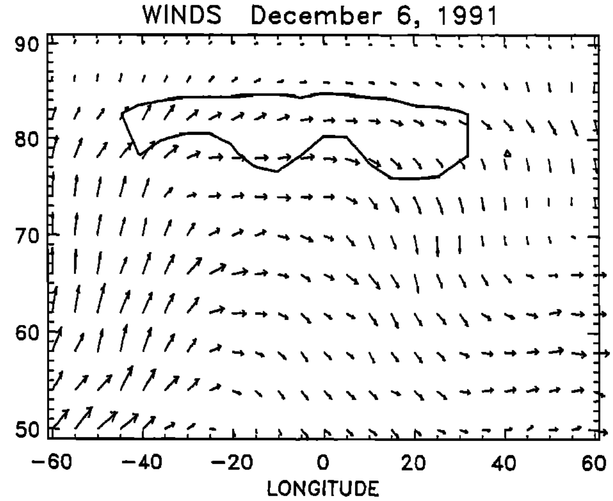

Fig. 3(a). The vector winds at 50 hPa for Dec. 6, 1991. The bold contour is the area where the temperature $T < T_c$ (criterion for heterogeneous processing).

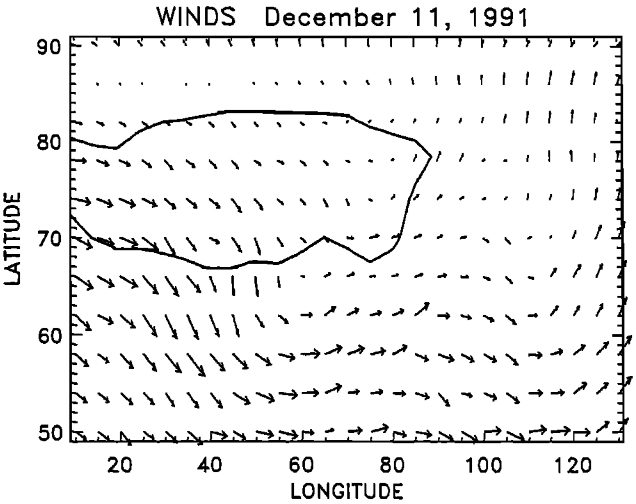

Fig. 3(b). Same as figure 3(a) but for Dec. 11, 1991.

Discussion

It is possible to characterize the behavior of these two events in terms of their dynamic structures. The vector wind fields associated with the two events are given in Figures 3(a) and 3(b). The area with $T \leq T_c$ is indicated by a bold contour. For both of these the zonal mean temperature is above the critical temperature for PSC formation. Values below the critical temperature are produced by rising and cooling associated with synoptic events. On Dec. 6, the area of cold temperatures is associated with an area of anticyclonic (clockwise) rotation imbedded in the cyclonic mean flow. The area of cold temperatures is found all along the northern edge of the anticyclone, which is consistent with rising and cooling throughout the anticyclone. On Dec. 11, the area of cold temperatures is associated with a region of cyclonic (counterclockwise) rotation imbedded in the mean flow. The cold temperatures appear at the western edge of the cyclone, consistent with sinking and warming at the center of the cyclone accompanied by rising and cooling at the edge. These differences produce the differences in whether the area of DELTA is spread or remains coherent. For the first case, the area of processed air is initially transported southward, consistent with the anticyclonic flow. As it reaches lower latitudes, influence of the cyclonic mean flow is seen, and the processed air is spread rapidly eastward. This is consistent with the behavior of the Jan. 31, 1989 ozone minihole observed by the Total Ozone Mapping Spectrometer and simulated with the 3D model [Rood et al., 1992]. The rising motion and cold temperatures accompanying an anticyclone lead to PSC formation and release of reactive chlorine; in the simulation there is rapid mixing of the processed air. In the second case, the processed air is transported slowly toward the north and towards the south. The area of cold temperatures and the area of processed air both move eastward. The cyclonic rotation of the feature within the cyclonic rotation of the vortex maintains an area of processed air that can be identified in the simulation for several days past those presented here, until processing from other events becomes significant.

Conclusion

Three dimensional simulations of processing by polar stratospheric clouds show the importance of mixing and transport to the distribution of reactive chlorine in the north polar vortex. This analysis indicates that for an anticyclone imbedded in the cyclonic polar vortex, processing produces a difference field DELTA which is subject to rapid shearing; a decrease in the maximum of DELTA is accompanied by growth of the area which is influenced by the processing event. In contrast, for a cyclone imbedded in the polar vortex, processing produces a difference field DELTA which maintains its maximum values longer and is spread much more slowly in the vortex. The evolution of the ClO as observed by MLS is in general agreement with the evolution of DELTA and the simple model of processing on PSC's without requiring significant photochemical conversion of reactive chlorine to reservoir species during this short integration. Both photochemical and transport processes should be considered in interpreting the ClO evolution associated with these two events.

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