QUALITATIVE STUDY OF THE BEHAVIOR OF MINOR SPECIES DURING A STRATOSPHERIC WARMING WITH A 3-D MODEL

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INTRODUCTION

It is well-known that the behavior of the ozone layer depends upon the coupling between several processes in the atmosphere. Natural or anthropogenic pollutants emitted from the surface or injected directly at high altitude may affect this layer. Assesment studies for long-term change of the ozone layer are conducted with the aid of various two-dimensional models (Brasseur et al., 1990; Garcia and Solomon, 1983,...). These models describe the long-term and seasonal evolution of minor constituents and take into account the interaction between all processes. However, many limitations affect the self-consistency of these models *e.g.*: the circulation in these models is only meridional and vertical and is not able to represent all types of motion in the atmosphere. During a perturbed winter in the north polar regions, the vortex is displaced from the pole to lower latitude so that wind may be reversed at a given location. Perturbed air masses are transported outside the darkened regions and may mix with local air masses (Mc Intyre and Palmer, 1984). Three-dimensional models are the only tools which can describe correctly these sporadic phenomena.

1. THE MODEL AND THE INITIALISATION

In this work, we use an improved version of three-dimensional model described by Ramaroson et al., 1990. The model results from the on-line coupling between a general circulation model (T21 version) (Cariolle et al., 1990) and a photochemical box model (Ramaroson et al., 1991). The photochemical package includes species of O_x, HO_x, NO_y, Cl_y and Br_x families and other source molecules *e.g.*: H_2O, N_2O, CH_4, CH_3B_r, CO, CFM's and CFC's. In the box model, no photochemical equilibrium is assumed. Ozone is interactive in the radiative codes of the GCM and is a prognostic variable in the box model. Photodissociation coefficients are computed using O_2 and O_3 as major absorbant. In the Schuman-Runge band, $J(O_2)$ is calculated with Frederick and Hudson method (1980). Rayleigh scattering and temperature dependence of cross-section of various species are taken into account. The four standard heterogeneous reactions occuring on PSC type I and II are taken into account. The condensation of HCl is parameterized as proposed by Marti and Mausberger (1991).

Two reactions in aqueous phase:

$N_2O_5 + H_2O \Rightarrow 2HNO_3$

$ClONO_2 + H_2O \Rightarrow HOCl + HNO_3$

using a background profile of aerosol are added to the photochemical system, the surface area of aerosol varies between $10^{-10}cm^{-1}$ to $5.10^{-9}cm^{-1}$ from 12 km to 25 km. CFC's and CFM's are fixed in the box from a zonal distribution computed by a two-dimensional model (Brasseur et al., 1990). Thirty-one species are transported independently. No family approximations are made and full diurnal variations are calculated.

The model initialisation is done with the analysis of ECMWF for January 17, 1987. The initial distributions of all species are derived from the 2-D model. Quantitative results depend strongly on these initial conditions especially for long-lived species. The aim of the study is to analyze the behavior of various species. Integration are carried out over 15 days. For all stratospheric levels, the vortex is lightly symmetric and centered at the North pole.

2. RESULTS AND DISCUSSION

Fig. 1a shows the distribution of the calculated PV on January 19 at a potential temperature surface of 1390 K (close to 3 hpa, 40 km). A tongue of air with relatively low values of PV is transported over the polar region. A high

value of PV, associated with the vortex is now located at 70°N-10°E with a tongue spread over the North-America. On Fig. 1b, 2 days later, apparently no entire "breaking" occurs over the pole but the PV value is almost constant (20.7 10^{-4} USI).

Due to the number of species studied in this work, all results cannot be shown here. In order to discuss, with a preliminary approach, the dynamical behavior of the various species, we have plotted the distribution of the variables at a specific universal time (12 UT).

The NO_2 behavior during a stable winter has been described by Ramaroson et al. (1990). When darkness prevails, NO_2 is converted via NO_3 to N_2O_5. A minimum of the NO_2 mixing ratio is computed over the North pole associated with sharp gradients due to the dependence of the lifetime versus latitude. For the perturbed winter we are studying here, Fig. 2a illustrates the NO_2 behavior (January 19). The incursion over the pole of high NO_2 air masses modifies the non-perturbed situation. The NO_2 minimum is found over high PV regions, in the vortex. After 2 days the behavior of NO_2 is changed. Fig. 2b shows a high value (14 ppb), originating from lower latitude, and transported over polar regions, is now surrounded by a tongue of low NO_2 (7 ppb). (The minimum calculated for the January 21 is 2 ppb). This situation may be easily interpreted with the help of the geopotential field maps for 20 and 21 January (not shown). After 11 days, Fig. 2c indicates that the mixing over North America is finished and chemical relaxation is complete. Another incursion of low latitude NO_2 occurs at about 110°E-20 to 40°N. The corresponding geopotential field is indicated on Fig. 3 on which one can notice a rapid incursion of air from low latitude (from Europ).

At mid-latitudes chemical lifetime for ClO is short. ClO mixing ratio varies with the solar zenith angle at all levels. At 40 km, it decreases slowly at night, to reach a minimum before sunrise; it increases rapidly when the sun rises. The main losses for ClO is the reactions with NO, O, and NO_2. The first two reactions are more rapid than the last one (only a few days). On Fig. 4a, we can see that the air incursion, mentioned above, gives low ClO mixing ratio. Two cells with high mixing ratio are calculated, associated with low NO_2 mixing ratio inside the vortex (see Fig. 2a). At mid-latitude, diurnal variations are not disrupted by this dynamical dependence. Fig. 4b gives the ClO distribution corresponding to Fig. 2c for NO_2. The first minimum over North America is quasi-steady (intermediate results not shown here). The second minimum located at 60°N-120°E is a dynamical effect, coupled with chemistry (see Fig. 3). The large maximum is also quasi-steady and corresponds to the position of the vortex.

Fig. 5 shows the cross-section for HNO_3 (condensed and gas phase). The maximum at 60°N (day) - 35 hPa is due to the condensation of HNO_3 (NAT formation) and conversion of $ClONO_2$ to Cl_2 and HNO_3 (solid) by reacting with HCl on the surface.

Fig. 6 illustrates the effects of PSC type I and II on the enhancement of ClO_x level (located approximately at 60N - 35hPa - > 0.4 ppb) by heterogeneous reactions. In the troposphere, cells of high ClO_x are due to reaction in aqueous phase which convert $ClONO_2$ to ClO_x.

The distribution of the temperature in Fig. 7 shows a strong cooling (about 185 k) at 60°N (day) - 35 hPa which occurs only during 3 days.

The behavior of HCl the 25 of january (Fig. 8) looks similar to that of HNO_3. However the main HCl production comes through the reaction with methane (CH_4 + Cl => HCl + CH_3), which is highly dependent on the temperature. At about 60°N 10 hpa (Fig. 8), the maximum of HCl (1.7 ppb) is associated with the incursion of CH_4 (Fig. 9) and high temperature (not shown but due to the warming about 246 K) At high latitudes, the reaction between HCl and OH is less effective due to the low concentration of OH. The inverse situation appears at 45°N. At these levels zonal winds are weak, the waves are broken and mixing may occur (Fig. 10).

CONCLUSION

Coupling between dynamics and chemistry is essential to understand the behavior of the different species. The 3D model which has been used in this work clearly shows the association of high vorticity with low NO_2, high HNO_3, HCl and ClO and a probable mixing during a wave breaking. The "normal" behavior during a stable winter is disrupted and high values of ClO and HNO_3 are transported with the vortex.

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Figure 1a

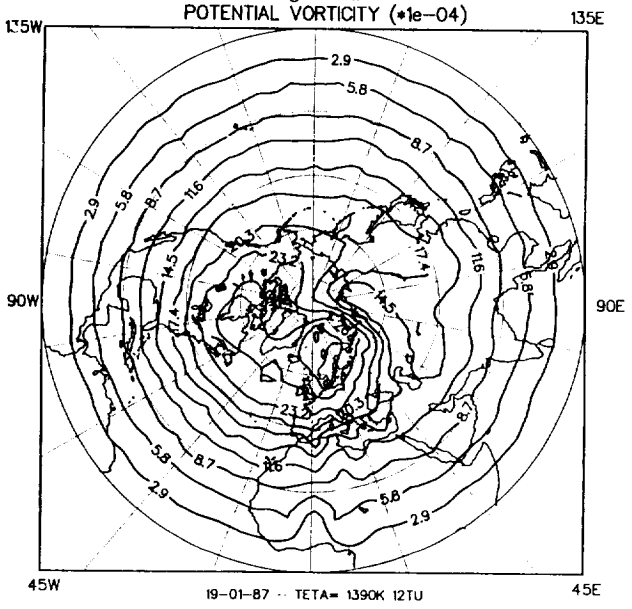
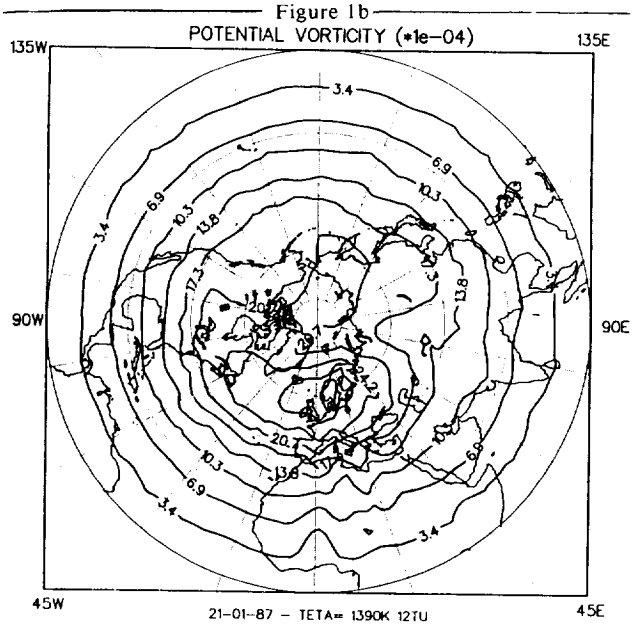
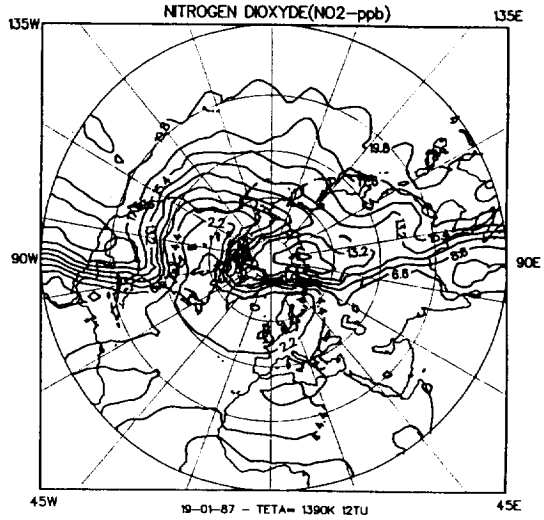


Figure 1b





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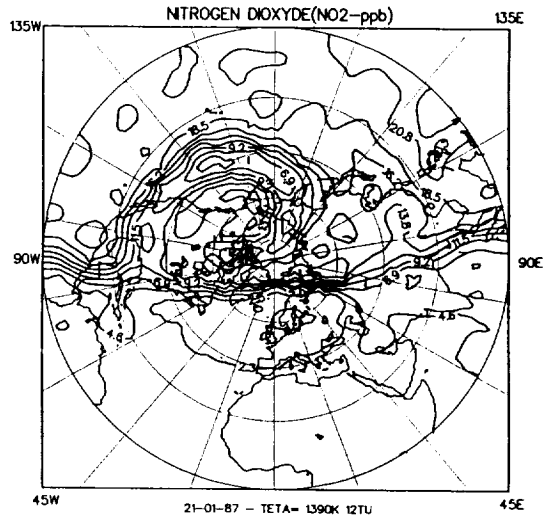

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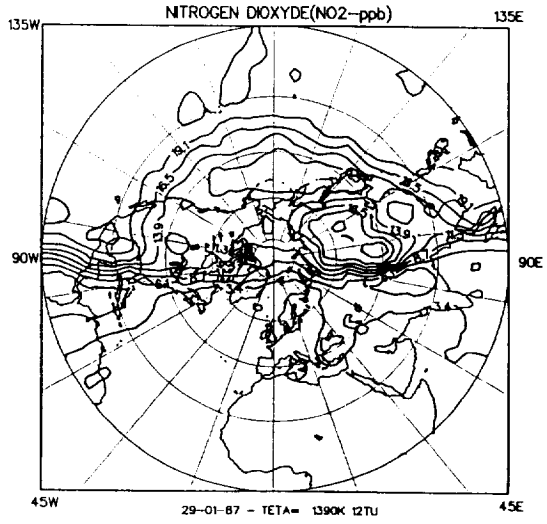

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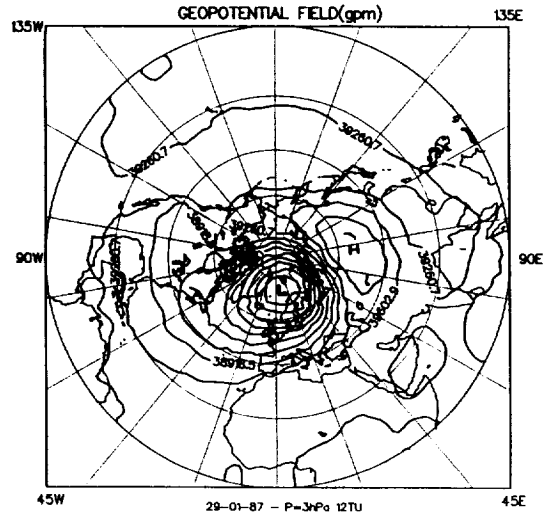

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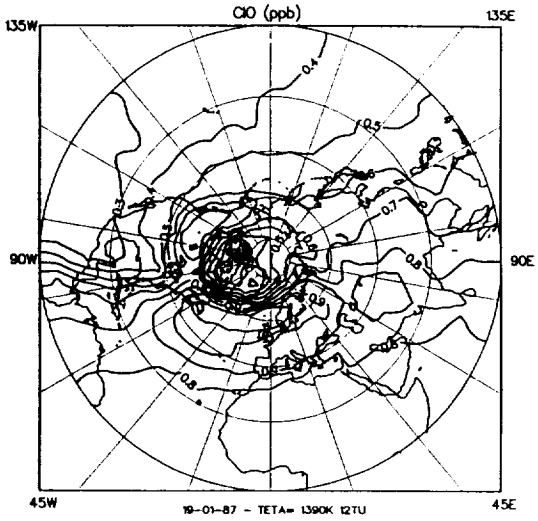

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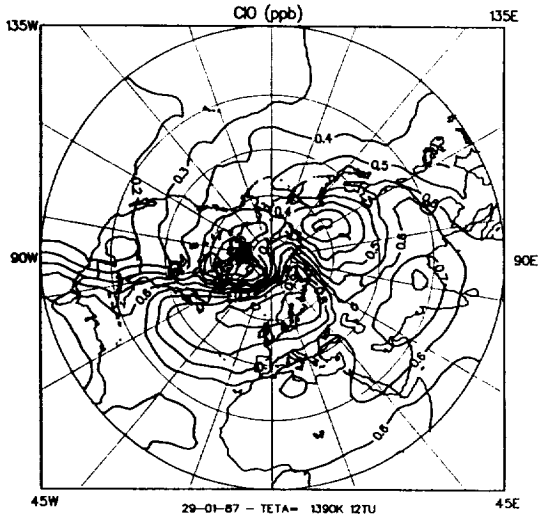

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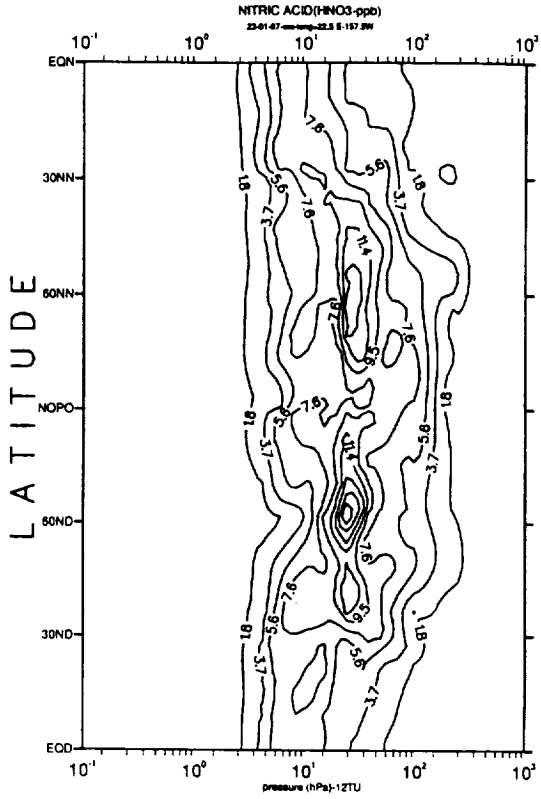

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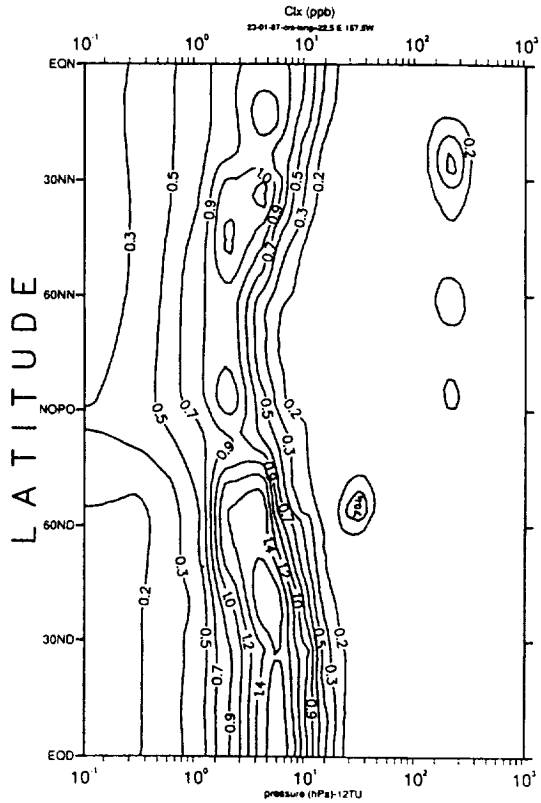

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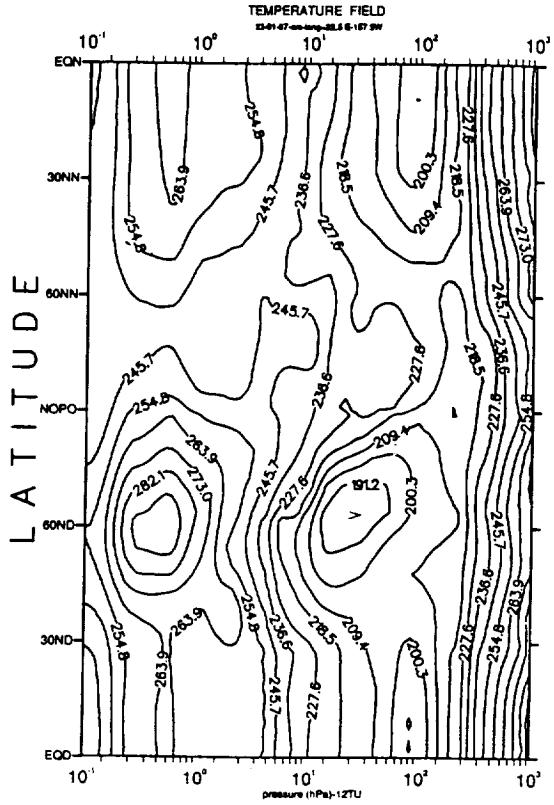

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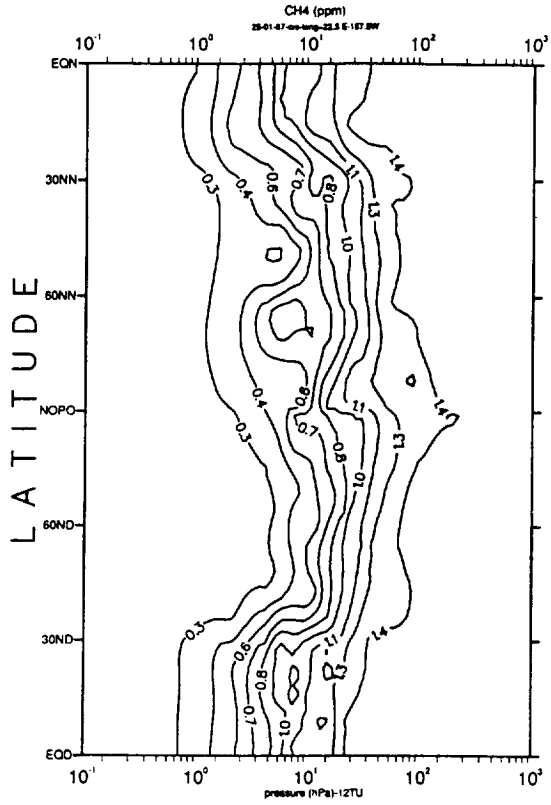

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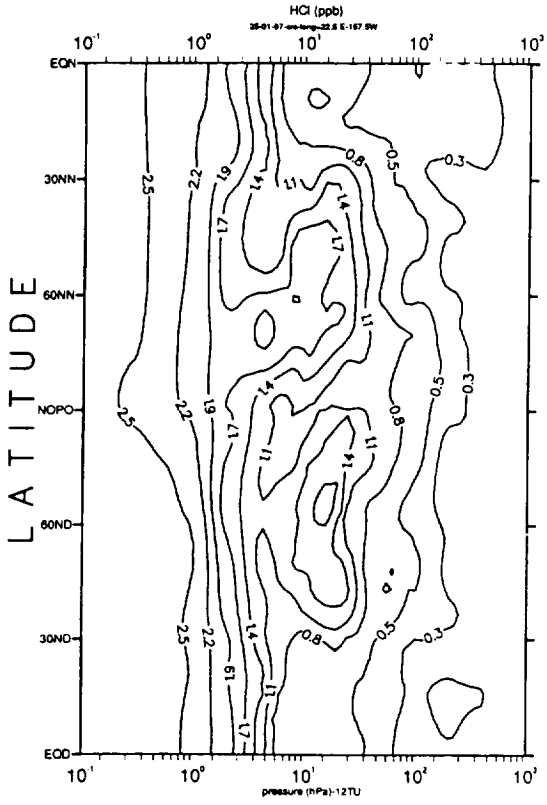

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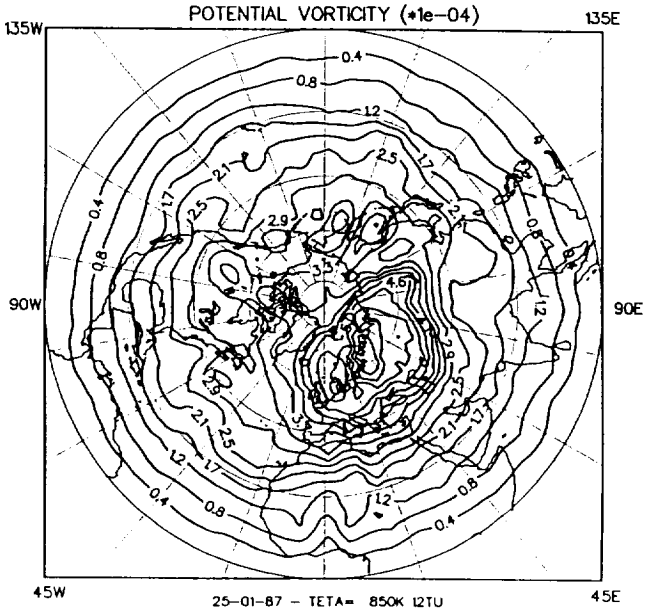

Figure 10