Impact of Physical & Chemical Parameters on Polar Ozone Concentration

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Submitted by

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DECLARATION

I hereby declare that the project work entitled ("Impact of Physical & Chemical Parameters on Polar Ozone Concentration") is an authentic record of my own work carried out at National Center of Antarctic & Ocean Research, Goa as requirements of one-year project internship for the award of degree of MTech in Environmental Science & Technology, Thapar University, Patiala, under the guidance of (Dr. K. Satheesan) and (Dr. Tapas Karmaker & Dr. N. Tejo Prakash), during June 2015 to June 2016.

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ABSTRACT

An analysis of stratospheric composition in past has revealed tight correlations in between multiple tracer gases. This correlation is a result of the same transport mechanism to which tropospheric gases are subjected to while being transported to the stratosphere through meridional circulation. This study attempts to find out the correlation of one such tracer i.e. ozone with other gases, known to have a certain degree of influence over ozone concentration in the stratospheric layer of atmosphere. This study also takes advantage of correlations obtained to predict future values of ozone volume mixing ratio in the stratosphere. This study has been carried out by using advanced machine learning tools which have been proven in past to give very accurate quantitative results. Such a study gives an insight into how chemical parameters work in synchrony under the background of climatological fields and influence the indispensable stratospheric ozone layer. This study also intends to show the influence of climatological fields of several variables that are derived during periods of ozone index amplification and during its weakening on ozone concentration. The premise here is that there are several robust supporting meteorological fields in the troposphere and in the stratosphere that go hand in hand with the accepted role of the chlorofluorocarbons that are attributed to the growth of the ozone hole and the implication is that however, these chlorofluorocarbons are necessary but not sufficient for the production of the ozone hole, that is supported by favorable meteorological fields. During periods of unfavorable meteorology, the breakdown of the ozone does not occur. For the purpose of carrying out this study, we use the reanalysis dataset NCEP/NCAR Reanalysis I for a duration of 41 years obtained from the archive of Physical Science division, Earth System Research Laboratory, NOAA. Polar meridional cell was one of the prominent variable examined in the process. The time tendencies of the ozone volume mixing ratio and of the intensity of the polar cell were calculated and used to identify those months during which there was a simultaneous rise or simultaneous decline in the time rate of change for both ozone gas volume mixing ratio and polar cell intensity. To further the study four cases were taken into consideration. Firstly, those months were considered during which both ozone and polar cell values were falling, thereafter those in which ozone was falling but polar cell value was rising and vice-versa, and lastly those months in which both were falling, as a function of time in all the cases. Sorting the climatology for these

four separate possibilities, respective climatologies were constructed. For the given four cases, aforementioned, climatologies of variables apart from ozone volume mixing ratio and polar cell were also considered. These comprised of the polar jet stream and the subtropical jet stream, the 50 hPa level air temperatures and the convergence of flux of temperature at that level. Finally, the results of this study exhibited robust differences in the atmospheric fields during the formation and weakening phases of the ozone hole, which was subjected to further analysis to find their influence, if any, over polar ozone layer.

CHAPTER 1 - INTRODUCTION

1.1 The Essence of Ozone Layer and A Short History of the Study On Its Depletion

The essence of stratospheric ozone layer has been well-established in previous literatures. The particularly sensitive ozone layer over the poles has been found to show spring associated ozone depletion occurring as a result of chemical molecules and physical parameters. Ozone (O_3) comprises a very small portion of the earth's atmosphere nearly 90 % of which resides higher in the stratospheric layer, approximately between 10 and 40 kilometers above the Earth's surface. This stratospheric ozone layer has been found to play a key role in filtering the UV-B region of electromagnetic spectrum of solar radiations. The unrestricted influx of solar UV-B radiations can have lethal impacts on not just humans but also on other fellow creatures and vegetation. Ozone in the stratosphere can absorb all of UV-C, some portion of UV-B and none of UV-A regions of electromagnetic spectrum respectively. Though UV-A and UV-C have their own set of impacts but the reason of major concern over here is the UV-B region of the solar electromagnetic spectrum. Prolonged exposure to UV-B radiations can result in a multiple ailments and disorders such as skin cancer, premature aging of the skin, cataracts, retinal damage so on and so-forth. Apart from this another important role played by ozone layer is its significant influence over atmospheric circulation and eventually over climate as well.

The depletion of this ozone layer occurs as a combined effect of the chemical reactions between a variety of reactive chemical molecules such as ClO- with hydrochloric acid, nitric acid *etc* under the influence of physical parameters such wind, temperature, solar radiation and precipitation. Halogen source gases which after being released into the troposphere are transformed into reactive halogen gases after reaching into the stratosphere. These halogen source gases the ones which participate in ozone depletion. Almost all of them now come under the vigilance of the *Montreal Protocol on substances that deplete ozone layer*.

Previous studies have shown the impact of ozone depletion on climate e.g. (*Rex et.al., 2004*). Ozone layer in the upper stratospheric region interferes with the solar influx/out flux, which in turn affects the atmospheric and ocean dynamics (*Solomon, 1999*). This is why, it is important to understand the causes behind its depletion, as they closely match with the pattern of climatological variations.

The Antarctic Airborne Ozone Experiment (AAOE) conducted in 1987, was first one of its kind, which had confirmed that chemical molecules such as chlorofluorocarbons and other sources of chlorine and bromine radicals control the rate of ozone depletion in the Antarctic to a large extent.

Also, *Chapman* had proposed that the ozone gas is formed by the photolysis of atmospheric oxygen. Oxygen is a very stable molecule, to the contrary the ozone produced in this process is highly unstable in nature. The atomic oxygen produced in the process reacts, with molecular oxygen to form ozone in the presence of a third body M, which stabilizes the given chemical reaction such as NOx, the need for which has been mentioned above. Solar energy (*hv*) is required for the reaction to occur. The bonds in the molecule of ozone are weaker compared to the bonds in molecular oxygen, hence ozone easily and quickly photolyzes again to form molecular oxygen (*Chapman, 1930*). Chemically balanced stoichiometric equation of the process have been given as below:

$$O2 + hv \rightarrow O + O \tag{1}$$

$$O + O2 + M \rightarrow O3 + M \tag{2}$$

$$O3 + hv \rightarrow O + O2 \tag{3}$$

$$O + O3 \rightarrow O2 + O2 \tag{4}$$

$$O + O + M \rightarrow O2 + M \tag{5}$$

Adding on to the fundamental discovery by Chapman, in 1974, *Rowland and Molina* had proposed a mechanism for photolytic destruction of ozone by chlorine atoms. Popularly being referred to as an extensive chain reaction, the theory proposed was as follows. During the austral season, the solar radiations of higher frequency would contribute to production of single chlorine atoms from reactive chlorine molecules. The mono-chlorine atoms would then combine with the stratospheric ozone molecules to form Chlorine-monoxide and molecular oxygen, which would then react with another ozone molecule to form chlorine atom and molecular oxygen. This process would result in the production of chlorine atoms, which would be available to react with other ozone molecules (*Molina and Rowland, 1974*). Hence, in this way, very few atoms of chlorine could contribute to a very significant amount of ozone depletion.

$$Cl + O_3 \rightarrow ClO + O_2 \qquad (6)$$

$$ClO + O_3 \rightarrow Cl + 2O_2 \qquad (7)$$

1.2 Ozone Depletion

Ozone depletion occurs as a combined effect of chemical reactions between a variety of chemical molecules, under the influence of physical parameters like wind, temperature, solar radiation and precipitation. Halogen source gases which are emitted into troposphere are converted into reactive halogen gases after reaching into the stratosphere. These halogen source gases are the ozone depleting substances and come under the vigilance of Montreal Protocol on substances that deplete ozone layer. The following text highlights the main breakthroughs in the ozone depletion studied over a long period of time.

1.2.1 Chapman's Mechanism for Ozone Production & Destruction

Chapman proposed that ozone is formed by the photolysis of atmospheric oxygen. The atomic oxygen produced then reacts with molecular oxygen to form ozone in the presence of a third body *M*, which stabilizes the given chemical reaction such as NOx. The solar energy (*hv*) is required for the reaction to occur. The bonds in ozone are weaker compared to the bonds in molecular oxygen, ozone photolyzed again to form molecular oxygen (*Chapman, 1930*).

The equation for the reactions described by Chapman has been given below:

$$O_2 + hv \rightarrow O + O \qquad (8)$$

$$O + O_2 + M \rightarrow O_3 + M \qquad (9)$$

$$O_3 + hv \rightarrow O + O_2 \qquad (10)$$

$$O+O_3 \rightarrow O_2 + O_2 \quad (11)$$

$$O+O+M \rightarrow O_2 + M \quad (12)$$

1.2.2 Rowland & Molina's Contribution

In 1974, Rowland and Molina had proposed the mechanism of photolytic destruction of ozone by chlorine atoms. Referred to as an extensive chain reaction, single chlorine atoms would combine with stratospheric ozone to form chlorine monoxide and molecular oxygen, which would react with another ozone molecule to form chlorine atom and molecular oxygen. This chlorine atom would now be available to react with other ozone molecules (*Molina and Rowland, 1974*) as given below:

$$Cl + O_3 \rightarrow ClO + O_2 \quad (13)$$

$$ClO + O_3 \rightarrow Cl + 2O_2 \quad (14)$$

1.3 Tracer Correlations

Tracer correlations studies has a lot of advantages. For one, tracer correlations provide an instantaneous climatology and help to minimizes the effect of small-scale geophysical variation that is generally observed in-situ datasets. For datasets of different sampling volumes, it provides a meaningful way of comparison, spatial and temporal resolutions. They also relax the coincidence criteria which is necessary when making direct comparisons. In reality such studies are done by various methods, one of which includes preparing correlation plots of one species against the other. The concept behind this method is called as "Equivalent displacement length". According to the concept of "Equivalent displacement length" in a given air sample the altitude deviations from mean profile of each individual species are similar to one another.

For majority of the tracer species found in the stratosphere there is no direct chemical correlation. But they tend to exhibit a strong correlation due a series of chemical and physical phenomenon. Majority of these species are produced in the stratosphere by photochemical reactions, predominantly in the tropical regions as the intensity of ultraviolet radiation is the maximum there, and majority of them tend to have a long life time. Tracer species can also be transported post their production the troposphere. Such species usually tend to remain inactive in the troposphere and also tend to have longer lifetimes (*such as chlorofluorocarbons*). They tend to exhibit correlations because they have long lifetimes and common regions of origin. *i.e* the stratosphere. Thus they are a subject to same transport and mixing processes. Following their production. Due to this, they tend to exhibit similar vertical gradients for a given range of altitude in the stratosphere. The correlations exhibited may not always be linear. This is because different gases may have different regions of maximum production/destruction in the stratosphere. [1]

1.4 Fuzzy Logic

Fuzzy logic is used in those instances when imprecision can be caused due the absence of a sharp criteria. It deals with the imprecision of facts. Fuzzy logic is based on the Fuzzy set theory which was given by Zadeh in 1965. According to the fuzzy set theory if we consider a universal set X, and a fuzzy set A within this universal set, then the relation between the fuzzy set, universal set and an element x belonging to the fuzzy set can be expressed by

$$A = \{x, \mu A(x) | x \in X\} \tag{9}$$

Where, μA represents the membership function of the element x to the subset A. The value of μA varies between 0 and 1. The closer is the value of μA to 1, more the element x belongs to fuzzy set A.

The basic building blocks of fuzzy logic are fuzzy sets, fuzzy operators. Fuzzy rules are also an integral part of the application of fuzzy logic. Fuzzy rules have two parts, the antecedent part and the consequent part. These rules are made up of IF and THEN statements (Takagi-Sugeno type). The part between IF and THEN is the antecedent part, this is the part where the inputs are defined, whereas the part after THEN is called the consequent part where the specifications for the outputs are given.

Fuzzy logic is applied to a problem, through a Fuzzy Interface System (FIS). There is a defined protocol which is followed while applying the fuzzy logic to a system. As a first step inputs are given to the FIS and their degree of connection to a given fuzzy set is estimated by fuzzy membership function. These inputs are crisp numerical values. Fuzzification - In this step these crisp numerical values are transformed to into linguistic terms (fuzzy sets). The Fuzzification layer in FIS generates membership values for all the numerical inputs using the membership function which was defined in the antecedent part of the fuzzy rule. The fuzzy logic controller in the FIS combines all the membership values to get what is called as firing strength. The next step now is to calculate values of consequents for each rule using the value of firing strength. The inputs are first classified based on their dynamic range as low medium and high. Using fuzzy rules (IF/THEN statements), the outputs are mapped to inputs.

1.5 Subtractive Clustering

If fuzzy logic has to be used to solve a given problem, fuzzy rules need to be generated. The number of fuzzy rules generated depend on the number of data points present in the feature space. If that is the case, the number of fuzzy rules will be very high and this will increase computational cost and time. One way to resolve this problem is to divide the data into different groups or clusters. This can be done by using a clustering algorithm. Clustering algorithms in turn can be of two kinds, supervised and unsupervised. A supervised clustering algorithm is the one in which the number of output clusters are predefined by the user. Unsupervised clustering algorithm to the contrary divide data into clusters without the help any predefined user specifications. Subtractive clustering algorithm is one such technique. The technique is very simple; the entire spectrum of data is considered as a data field. Two key elements are involved, cluster center and cluster radius. A cluster center is a point with the maximum number of points surrounding it. Once a cluster is created it is removed from the feature space to avoid the presence of same points in another clusters. One by one the entire dataset is grouped into clusters. The number of clusters generated depends upon the value of cluster radius chosen.

Mathematically, cluster radius is the Euclidean distance between cluster center and its surrounding points. Cluster radius plays a crucial role in deciding the model efficiency. If the cluster radius is small the number of clusters will be more, but this will lead to overfitting. To the contrary, if the cluster radius is large, it may reduce the model accuracy.

Hence, a trial and error chosen value of cluster radius which maintains a balance between fitting and accuracy has to be chosen. Apart from reducing the rule base, subtracting clustering also exhibits automatic rule generation capability.

1.6 The Geography of Southern Poles of Antarctica

Southern Polar region or Antarctica is surrounded by water from all its sides, which isolates it from land masses up to a large extent. This seclusion from land masses results in a unique meteorology of the southern poles and in much more extreme weather conditions than northern poles. A very important feature of this extreme weather in the southern poles is the infamous polar vortex which again is an isolated mass of air swirling over the poles. Precisely, it's a large area of low pressure region. It exists near the both the poles. During the polar winters, when the air temperatures drop to as low -90 degree Celsius, the naturally occurring gases such as water vapor and nitric acid present in the stratosphere begin to condense out, leading to the formation of ice crystals. These ice-crystals contribute to the formation of Polar Stratospheric Clouds, which have been associated with the springtime depletion of ozone layer for time immemorial. These polar stratospheric clouds are of different types depending on their chemical composition. These are namely PSC I and PSC II, which can be further divided into PSC I a & b, and PSC II. These polar stratospheric clouds provide a solid surface for certain heterogeneous reactions to occur. These chemical clouds do not participate in the reaction, but instead provide a solid surface for this reaction which reduces the threshold energy of activation required for the reaction and results in a faster rate of chemical reaction. These reactions which contribute to the destruction of ozone in the Polar Regions have been stated below:

$$HCl + ClONO_2 \rightarrow HNO_3 + Cl_2 \qquad (15)$$

$$ClONO_2 + H2O \rightarrow HNO_3 + HOCl \qquad (16)$$

Through these kinds of reactions, reservoir species of chlorine are converted into more reactive species. However, there is another side to the polar stratospheric clouds as well.

Apart from providing a surface for the above reaction to occur, they also play a role in the removal of gaseous such as nitric acid. This gas would have otherwise combined with chlorine monoxide to form chlorine nitrate which would be less reactive and thus less harmful to stratosphere.

Adding on to this, dehydration and denitrification due to and in this clouds, also play a major role in controlling the rate and magnitude of ozone destruction over poles. During polar winters the size of polar stratospheric clouds increases gradually with more and more freezing of Nitric acid and Sulphuric acid. They tend to become heavier and precipitate out; this is referred to as denitrification of stratosphere. If denitrification occurs lesser nitric acid is available to quench or deactivate reactive chlorine, which would lead to more ozone depletion. But if intense denitrification occurs during early polar winters, then it helps suppress the formation of polar stratospheric clouds leading to lesser ozone depletion.

The study on impacts of ozone depletion over climate has been carried out for a long period of time. In one such study by *John Turner* and others in the year 2009[1], a group of scientist had studied the impact of stratospheric ozone depletion over sea ice extent and sea level rise, through changes in cyclonic circulation over the southern poles, via a chain of reactions starting from ozone depletion. In another study by *Wenju Cai and Tim Cowan,* published in a journal of AMS (American Meteorological Society) ozone depletion has been confirmed to have an impact on subtropical gyre circulation [2]. Apart from modulating the existing circulation parameters ozone has also been found to have an impact on the temperature trends over polar regions, both north and south. In a study carried out by *William J. Randel and Fei Wu* [3] using satellite data of air temperature and ozone volume mixing ratio, the spring associated ozone depletion has been observed to result in a significant cooling of the polar regions, this data is consistent with model predictions which include the forcings of ozone depletion and radiosonde observations. Another paper by Kang and others outlines the importance of the impact of polar ozone depletion on subtropical precipitation [4]. A different study, has showed the impact of changes in water vapor over ozone concentration through changes in climate parameters. The study indicates that the changes in water vapor concentration brought about by the variations in sea surface temperature result in modulations in the polar vortex, intensifying it and leading to enhanced ozone depletion [5]. The study on impacts of ozone depletion over climate have been carried out for a long period of time. In one such study by *John Turner* and othersin the year 2009[1], a group of scientist had studied the impact of stratospheric ozone depletion over sea ice extent and sea level rise, through changes in cyclonic circulation over the southern poles, via a chain of reactions starting from ozone depletion. In another study by *Wenju Cai and Tim Cowan,* published in a journal of AMS (American Meteorological Society) ozone depletion has been confirmed to have an impact on subtropical gyre circulation [2]. Apart from modulating the existing circulation parameters, ozone has also been found to have an impact on the temperature trends over polar regions, both north and south. In a study carried out by *William J. Randel and Fei Wu* [3] using satellite data of air temperature and ozone volume mixing ratio, the spring associated ozone depletion has been observed to result in a significant cooling of the polar regions, this data is consistent with model predictions which include the forcings of ozone depletion and radiosonde observations. Another paper by *Kang* and others outlines the importance of the impact of polar ozone depletion on subtropical precipitation [4]. A different study, has showed the impact of changes in water vapor over ozone concentration through changes in climate parameters. The study indicates that the changes in water vapor concentration brought about by the variations in sea surface temperature result in modulations in the polar vortex, intensifying it and leading to enhanced ozone depletion [5]. *Proffitt et. al.* [6] conducted a study from 1991 to 1992 to estimate the ozone loss during winters in Northern Polar Vortex. They found the concentration to be increasing with the altitude and concluded that ozone loss within Polar Vortex was altitude dependent. The reason behind such an observation was found to be the impact of reactive chlorine on the ozone rich air which was descending down the vortex. As the air descended and reacted with reactive chlorine, it got depleted of ozone, thus the air which was finally released from the bottom of the vortex was significantly lower in ozone concentration. In a study by *Schoeberl et. al.*[7], it was found that the overall size of polar vortex had a large influence in deciding the areal coverage of ozone depletion. This explained the reason behind comparatively less ozone depletion in Northern Poles as compared to the Southern Poles due to the restricted region of maximum cold leading to a smaller areal coverage of polar vortex. In another study by *Polvani et. al.*[8] it was found that ozone depletion impacts not just the polar tropopause and the position of midlatitude, but it also results in broadening of Hadley cell and poleward extension of subtropical dry zones. The study concluded that Southern Hemisphere Tropospheric Circulation changes could largely be caused by polar stratospheric ozone depletion.

Jet streams are strong winds flowing near tropopause at around 160 km/h. They are 160 km wide, 2-3 km in thickness and several kilometers in length. These jet streams are formed at locations where there is a steep gradient in temperature. This usually occurs at two locations, one is the boundary between polar and midlatitude winds and the other one is the boundary between midlatitude and tropical winds. The former one is called as polar jet and the latter one is called as subtropical jet. Both these jets travel uniformly around the earth from west to east. These jet streams are influenced by the Coriolis effect and land masses, which in turn influence these jet streams through friction and the temperature difference. Coriolis effect makes these changes more prominent. With the increase in temperature gradient the strength of the polar jet increases and vice versa. Moreover, as the strength of the waves decreases the meandering of the waves decreases and vice-versa. Understanding of jet stream is very essential as the various attributes of jet stream decide the kind of influence it will have in regions it surrounds. Such as the position of jet stream decides the kind of weather which will be experience by that region. The direction and angle by which a jet stream arrives a given region decides what kind of air will be carried by jet air to that region.

Though previous studies have focused more on the impacts of climatological parameters over polar ozone concentration, this study in turn tries to map the impact of climate change over ozone, a topic which is understudied but of a very high significance. Through, this study we have tried to show various climatological fields for several variables that are derived during periods of ozone amplification as well as weakening try to establish their participation in ozone depletion in combination with chlorofluorocarbons.

CHAPTER 2 - LITERATURE REVIEW

Stratospheric ozone layer has been found to play a very important role in filtering UV-B radiations and preventing them from reaching earth`s surface. The uncontrolled solar input of UV-B radiations can have lethal impacts on not just human health but also vegetation. Ozone layer also exhibits a significant influence over the atmospheric circulation and thus over climate as well. Previous studies have shown the impact of ozone depletion on climate e.g. (Rex et. *al*., 2004). Ozone layer in the upper stratospheric region interferes with the solar input/radiation, which in turn affects the atmospheric and ocean dynamics (Solomon, 1999). This is why it is important to understand the causes behind its depletion The *Antarctic Airborne Ozone Experiment* (AAOE) conducted in 1987, first one of its kind, had confirmed that chemical molecules such as chlorofluorocarbons and other sources of chlorine and bromine radicals control the rate of ozone depletion in the Antarctic to a large extent. This was followed by the Airborne Arctic Stratospheric Expedition (AASE) in 1989. Vortex data from both the poles showed distinct variations, resulting in differences in the way and the extent of ozone depletion occurring there. While the Polar Vortex developing over the Antarctic in winter season exhibited desiccation along with depletion of NOy, minor changes in NOx/NOy ratios were observed over the Arctic Polar Vortex.

Modelling studies can be used for predicting future ozone losses by simulating the real time environment. This can be done by taking into account the influence of atmospheric circulation and the concentration of reactive species known to be involved in ozone depletion. Extensive studies have been undertaken in this area previously. Conventionally, chemistry–climate models are used in order to develop a deeper scientific understanding of the chemical and physical parameters involved in ozone depletion (Zeng et. *al.,* 1993; Austin, 2002). *Lemmen* et. *al (2006),* studied the chemical ozone loss in lower stratosphere using a chemistry climate model ECHAM4.L39 (DLR)/CHEM model. Making use of tracer correlations between ozone and methane they tried to understand the changes in ozone column over a period of 40 years from 1960-1999. However, the model failed to take into account the full contribution of chlorine loading and thus predicted only 63% of the ozone loss as shown by HALOE observations of ozone. Feng et. *al* (2010) reported the use of SLIMCAT an offline 3D Chemical Transport Model with a detailed stratospheric scheme and a NAT-based denitrification scheme to study the rate of ozone loss over Arctic stratosphere for years 1999/2000, 2000/2002 and 2003/2004. The study also made use of a CCM scheme to incorporate vertical transport from troposphere to stratosphere. The model was able to predict the rate of ozone loss more accurately and the results were in good agreement with the observational data.

Some of the previous studies have modelled the influence of greenhouse gases in bringing about tropospheric warming and stratospheric cooling, which promotes the formation of polar stratospheric clouds thus increasing the rate of ozone depletion. e.g. (Shindell et. *al,* 1998). Using a simple chemistry scheme as an input to the Global Circulation Model they were able to incorporate the influence of increasing GHGs over lower stratospheric temperatures and thus predict the increased rate of ozone depletion. Another group of scientists had tried to model the impact of changing concentration of ozone and carbon dioxide on temperature of stratospheric air (Fels et. *al, 1980*). To model the impact of a 50 % increase in carbon dioxide and a 50 % decrease in ozone on stratospheric temperature scientists had made use of Global Circulation models along with supporting radiative models. Shindell et. *al. (*2000*)* used the GISS Climate Atmosphere Model to study the impact of increased greenhouse gas concentration, polar ozone loss, solar cycle variability and volcanic eruptions on northern hemisphere winter climate. Though CCM`s are considered to be very efficient but they are computationally very expensive. They are data intensive and take a long time for estimating the final results. Hence, faster chemistry schemes are essentially needed for incorporating in the models, so that the computational efficiency can be increased (Rex *et. al*., 2014). Semi-empirical models have the advantage over CCM of a lesser run time. In 2005, Taylor et *al* had proposed a chemistry scheme called the Fast Stratospheric Ozone Chemistry (FASTOC) scheme which was three orders of magnitude faster than a stiff kinetic equations solver. When coupled to GCM it could successfully predict the rate of ozone loss in good agreement with observational data. Another semi empirical model called as SWIFT (Semi empirical Weighted Iterative Fit Technique) based on a set of coupled differential equations could estimate the rate of ozone loss in polar stratosphere taking the concentrations of a set of chemical parameters as input. (Rex et. *al.,* 2013).

Tracer interrelationships between chemical molecules are widely used in estimating the concentrations of one chemical against the other. Satheesan (2012) have explained the use of tracer correlations in generating long-term time-series data of tracers which is otherwise difficult to generate or acquire. Popp et. *al.* (2009) have explained on how tracer correlations can be used to generate long time profiles of nitric acid in the lower stratosphere using the widely available Ozonesonde data. Tracer interrelations have also found application in the calculation of ozone loss in Arctic winters by Tilmes et. *al*. (2003). Previous studies using tracer-tracer correlations show excellent examples of how tracer correlations can be used to study the causes behind various events occurring in the lower atmosphere which otherwise cannot be explained due to the unavailability of sufficient data set. As the spatio-temporal resolution of remotely sensed data and in-situ data usually vary significantly, tracer correlations provide an alternative for validating the satellite data. Tracer correlations provide instantaneous climatology by reducing the influence of small scale geophysical variations over the in-situ data or remote sounding data. The only significant chemistry been ozone and nitric acid occurs on the surface of polar stratospheric clouds where condensed nitric acid reacts with hydrogen chloride to form chlorine nitrate. A more significant connection that exists between these two species is a dynamic correlation. It exists because both ozone and nitric acid are photo-chemically produced in the same region i.e. the middle stratosphere and both are subjected to the same transport and mixing after formation (Murphy et.*al*., 1993). Being subjected to similar conditions their vertical profiles exhibit common features, explaining why strong correlations exist between ozone and nitric acid in the middle as well lower stratospheric region.

Apart from HNO3 - O3 correlations, HCl - O3 correlations have also been used in past to estimate the concentration of one against the other. Marcy et. *al.* (2004) had developed a methodology to estimate the concentration of ozone in upper tropospheric region by studying stratosphere to troposphere transport of air parcels and tracer correlations between ozone and hydrogen chloride. Ozone also shows dependency over physical parameters. Its dependency over temperature has been studied in past as well. Ozone-temperature dependency has not just been studied to find the influence of temperature over ozone but also to estimate the concentration of certain chlorine reservoirs in the stratosphere. These kinds of studies have taken into account that ozone temperature dependency gets affected in the presence of a chlorine reservoir (Stolarski et.*al.,* 2012). Usually such kinds of studies are done over a seasonal scale to consider a wide range of temperatures. Previous works have also tried to establish the dependency of temperature variations over stratospheric ozone columns (Yjun et. *al.,* 1993) and explain various atmospheric events as a result of that dependency (Petzoldt et. *al.,* 1994) Stratospheric ozone depletion also has an influence over precipitation. In a study conducted on the precipitation and ozone between 1979 and 2000, the subtropical precipitation in the Southern hemisphere has been found to increase with decreasing polar ozone concentrations. (Kang et. *al.,* 2007). Another study explained the influence of precipitation over ozone loss in the arctic region. Precipitation leading to denitrification renders the polar stratosphere devoid of Nitric Acid Trihydrate which can instead deactivate the reactive chlorine radicles and reduce the rate of ozone loss (Waibel et. *al.*, 1999). This explains the interdependency of ozone mixing ratio and precipitation over each other.

Brewer- Dobson Atmospheric circulation and polar ozone loss display a strong dependency over each other as well. Previous studies have not just reported the influence of atmospheric circulation over ozone mixing ratio in polar stratosphere. But also the influence of ozone depletion over the atmospheric circulation. Studies done in the latter half of twentieth century suggest that most of the changes in the tropospheric circulation over the Sothern Hemisphere have been brought about by polar ozone depletion. e.g. (Lorenzo et. *al*., 2010). This study aims to improvise over previously done studies by considering multiple parameters at one go instead of considering one parameter at a time. This helps in simulating the real-time environment more closely. As chemical parameters alone don’t influence the phenomenon of ozone depletion

(Solomon and Gracia, 1986), physical and chemical parameters both will be taken into consideration concurrently.

*Statistical analysis***,** a commonly followed technique for correlation analysis includes regression analysis. Least square regression analysis technique has been used in past to develop correlations between ozone and nitric acid. Popp et. *al.* had used the least square regression analysis to find out the correlation between ozone and nitric acid in the lower and middle stratosphere. The technique had also been used by Lowenstein and Podolske, 1993 to find NO2/NOy correlations in lower stratosphere. However, there are many drawbacks when we use simple linear relations based on regressions. More advanced techniques, which takes care of the non-linearity will be much better. Fuzzy logic is one such non-linear optimization method which can be used to study the tracer correlations in the lower stratosphere as compared to regression analysis studies (Satheesan, 2012). Tagaki and Sugeno *(1985)* have described the use of fuzzy logic for modelling purposes. In addition to fuzzy logic, genetic algorithm has also been gaining popularity in solving non-linear optimization problems as described by Gallagher & Sambridge (1994) and Loughlin, 2007 which are used for many modelling studies. Recently, Basu et. *al.*, (2005) used genetic algorithm for predicting wave heights in the North Indian Ocean.

The expected outcome of this study is a semi-empirical chemistry scheme which can be used for quantitative forecasting of the ozone values taking into consideration the influence of fundamental chemical parameters like HCl, HNO_3 and $ClONO_2$ and physical parameters such as temperature, wind, pressure etc.

2.1 Objective of the study

The objective of this study is to find out that how a range of climatological fields and chemical molecules influence the ozone layer found in polar stratospheric region, southern polar in this study. It maps and highlights the salient features of these meteorological fields which support the rapid buildup and decline of the ozone layer. It also gauges the effect of a variety reactive molecules of polar stratospheric ozone concentrations. The premise of this study is that although the presence of excessive reactive chlorofluorocarbon molecules in the lower to mid stratospheric region is necessary but it alone cannot contribute to the depletion of polar ozone layer. Hence, this work establishes the very essence of these climatological fields & chemical parameters in context to their role in changing ozone concentration over poles.

CHAPTER 3 - STUDY AREA &DATA COLLECTION

3.1 Data Used

JAXA (Japanese Aerospace Exploration Agency) operated ILAS-II (Improved Limb Atmospheric Spectrometer-II) data has been used for this study. The instrument provides vertical profiles of ozone and nitrous oxide along with many other trace gases with a vertical resolution of 1 kilometer. Measurements covering a latitudinal range of 54 degrees north and 71 degrees north and in the altitude region of 10-50 kms (stratosphere) have been used. The ILAS-II N2O data used is in agreement with the balloon values and Odin sub-millimeter radiometer within 20%, however this is applicable only when the volume mixing ratio of nitrous oxide is greater than 250 ppbv. When the volume mixing ratio is in between 30-50 and 250 ppbv, then the ILAS-II data is underestimated by 10-30%. In case of the ozone however, the data used is within 10% agreement with other datasets for altitudes between 11 and 40 kilometres, with a positive bias above 20 km and a negative bias above it. Beyond an altitude of 40 kilometres, the negative bias increases and reaches nearly 30% around 61-65 kilometres. The chosen altitude is suitable for quantitative data analyses in the given latitude region. As compared to the ILAS-II version 1.4, the ILAS-II version 2 data which has been used for this study, is better. The improvement can be attributed to transmittance correction in the northern hemisphere as well as the usage of an improved tangent height registration.

For the purpose of carrying out this research work, the reanalysis dataset from NOAA has been used. The data has been generated, archived and made freely available to the scientific community by Physical Sciences Division, Earth System Research Laboratory, NOAA (National Oceanic & Atmospheric Administration).

The NCEP/NCAR Reanalysis 1 projects uses a state-of-the-art analysis/forecast system to carry out data assimilation using the data available from 1948 to the present time. A major subset of this assimilated data is available from Physical Science Division of ESRL. This data was originally available with a temporal resolution of 4 times daily. Daily mean values of the dataset had also been computed and made available. The data which is available from 1948-1957 is non-Gaussian gridded data, which was done 8-times daily in the model. This is because the inputs were available at 3Z, 9Z, 15Z and 21Z, whereas the 4-times daily data has been available at different levels of 0Z, 6Z, 12Z, and 18Z. The combined result for the earlier period was obtained by forecasting the data for the missing levels. It is available in various formats, such as daily, 4-times daily and monthly means and is divided into different sections such as pressure levels, surface, surface flexes, tropopause, derived dataset and spectral coefficients. This study utilizes a subset of the available data set in terms of parameter, time period, hyper-parameters used etc. Such the wind dataset set from this database was available U wind and V wind, both of which were available in three different formats at 4 times daily, daily means and monthly means, both available from a time period of 1948 onwards till present time. Both the datasets cover a global spatial domain and were available over 17 pressure levels and 28 sigma levels, at a spatial resolution of 2.5 degrees making up to 144 by 73 grids in all. This was the standard format followed for all the datasets available in the archived database. However, for the purpose of this study we restricted our data usage to monthly mean values U-wind and Temperature datasets spanning over a time period of 41 years from 1974-2014, ranging over a latitudinal resolution of 60 degrees South to 90 degrees South, a longitudinal resolution of 0 Degree East to 357.5 Degree East, and over a range of pressure level from 10 hPa to 100 hPa. In all the dataset contained 144 by 36 grids (Southern Hemisphere Only). This dataset was subjected to multiple stages of processing and mathematical computation to finally obtain a dataset which can be used a graphical representation of the given parameter. This representation helped in getting insights into the raw data and draw conclusions about the physical parameter under observation.

A second set of important dataset was the precipitation dataset. This dataset was obtained from the NASA archives of TRMM, Tropical Rainfall Measuring Mission dataset. The Tropical Rainfall Measuring Mission (TRMM) is a joint mission between NASA and the Japan Aerospace Exploration (JAXA) Agency to study rainfall for weather and climate research. The satellite with a mission to collect global tropical rainfall and lightning data was launched in late November 1997 and it continued up to April 15, 2015. Although the equipments were designed to work for a period of 3 years, however they continued to work for a long period of 17 years. The satellite contained 5 instruments in all, one of which was a 3-sensor rainfall suite (Precipitation Radar, TRMM Microwave Image, Visible and

Infrared Scanner) and 2 related instruments (LIS and CERES). The data obtained through TRMM has been popularly used in projects related to tropical cyclone structure and evolution, convective system properties, lightning-storm relationships, climate and weather modeling, and human impacts on rainfall. Apart from the above mentioned research applications, it has also been found to have operational applications such as flood and drought monitoring and weather forecasting. For the given study, 3B43 dataset of TRMM was used. The purpose of 3B43 algorithm was to produce the best-estimate precipitation rate (in mm/hr) and root-mean-square (RMS) precipitation-error estimates from TRMM and other data sources. The algorithm used for generating the dataset made a combination of multiple independent estimates of precipitation TMI, Advanced Microwave Scanning Radiometer for Earth Observing Systems (AMSR-E), Special Sensor Microwave Imager (SSMI), Special Sensor Microwave Imager/Sounder (SSMIS), Advanced Microwave Sounding Unit (AMSU), Microwave Humidity Sounder (MHS), microwave-adjusted merged geo-infrared (IR), and monthly accumulated Global Precipitation Climatology Centre (GPCC) rain gauge analysis. All of the input microwave datasets were inter-calibrated to TRMM Combined Instrument (TCI) precipitation estimates (TRMM product 3B31); the iIR estimates were computed using monthly matched microwave-IR histogram matching; then missing data in an individual 3-hourly merged-microwave fields were filled with the IR estimates. After the completion of preprocessing, the 3-hourly multi-satellite fields were summed for the month and combined with the monthly gauge analysis using inverse-error-variance weighting to form the best-estimate precipitation rate and RMS precipitation-error estimates. These gridded estimates had a monthly temporal resolution and a 0.25-degree by 0.25-degree spatial resolution. Spatial coverage extends from 50 degrees south to 50 Degrees north latitude. For the purpose of this study the spatial domain of the dataset used was restricted to 30 degrees by 30 degrees.

3.2 Study Area

The continent of Antarctica is centered asymmetrically around the southern poles. It spans an area of nearly 14 million kilometer squares and 98 percent of this area is covered by ice-sheets. It is surrounded from all the directions by Southern Ocean waters or more precisely the southern waters of Pacific, Atlantic and Indian Oceans. It is the coldest, driest and windiest place on the earth where temperatures fall to as low as approximately -90 degree

Celsius during winters. The annual precipitation rates in Antarctica go only up to 200 millimeters annually, thus making the Antarctic land a dry and a deserted area. There are multiple reasons behind the southern pole being colder than the northern pole. Firstly, as attributed to the high elevation of much of Antarctic continent (Greater than 3000 meters above the mean sea level), the tropospheric inverse lapse rate plays a predominant role in Antarctic cooling. Secondly, during south-polar winters which fall in July, earth is farthest from the sun (this point on earth's orbit is called aphelion) and during south-polar summers which fall in January, earth is closest to the sun (this point on earth's orbit is called perihelion). This leads to a colder Antarctic winter. Apart from the above mentioned facts, the popular Katabatic wind determine the near surface wind fields of Antarctica. These are high speed winds which descend down the hill or high slopes of Antarctica, once they become dense enough due to cooling.

During the polar nights when heat is radiated by elevated surfaces, the air in contact with these surfaces also cools down and begins to descend under the influence of gravity, giving rise to Katabatic winds. Generally, as the air descends its temperatures raises due to compression, but in the case of Katabatic winds blowing over Antarctica, the air is still intensely cold when it reaches the ground, thus keeping the near surface temperatures very low.

Another important feature of the polar climatology is the polar vortex. A polar vortex is a pattern of winds which resemble cyclones. The base of this vortex lies in the middle and upper troposphere and extends upto the stratosphere. They are formed when the stratospheric air swirls above the poles in circular motion during polar winters, with its center containing relatively still air. They form a shield around the poles and prevent the warm outside air from mixing in. When the polar vortex weakens, then the temperatures within these polar vortices go as low as -90 degree Celsius. Such temperatures are favorable for the formation of the infamous polar stratospheric clouds, which are responsible for the spring associated ozone depletion in southern poles.

These polar stratospheric clouds (PSCs) are also known as nacreous clouds (nacre means mother of pearls). These are colorful clouds which are formed in an altitude range of 15 to 25 kilometres.

PSCs are of two types, Type I and Type II. Type I is formed at temperatures nearing -90 degree Celsius (~85℃). Type II however, are formed at relatively higher temperature of nearly -80 degree Celsius (~78℃). They are again classified as Type Ia, Type Ib and Type Ic. Type Ia contains crystalline compounds of water and nitric acid, such as Nitric Acid Trihydrate (NAT). Type Ib contains droplets of solutions of sulphuric and nitric acid. Type Ic contains metastable nitric acid-water phase. Type I PSCs basically serve as a medium for heterogeneous gas phase chemical reactions to occur at higher rate than would have been possible without a solid medium leading to a rapid ozone depletion during spring season. On the surface of polar stratospheric clouds, two sets of reaction occur. In the first set, the reservoir species of chlorine combine to release molecular chlorine. In the second set, the molecules which can restabilize the chlorine species (which were previously bonded with reservoir species of chlorine) get adhered to the surface of PSCs. As the winter progresses, and the polar stratospheric clouds become denser, they start to precipitate out, leaving the molecular chlorine species up in the stratospheric air, and bringing with them the molecules which can quench the reactive species of chlorine. Now as the spring hits the polar regions, high energy photons of the ultraviolet radiations from the sun split the chlorine molecules into chlorine atoms. These chlorine atoms then react with the weakly bonded oxygen atoms in ozone and break down the ozone into oxygen atoms and an oxygen molecule. They combine with the oxygen molecules to form chlorine monoxide. Two such chlorine monoxide molecules combine to release a chlorine molecule and an oxygen molecule. This chlorine molecule is again splitted into chlorine atoms and it is now free to deplete the ozone layer further.

CHAPTER 4 - RESEARCH METHODOLOGY

4.1 Estimation of Polar Stream Function Using Wind Data

Mean meridional Circulation is a zonally symmetric motion. The mean meridional circulation is usually depicted via the use of a stream function. For zonally symmetric motions, stream function is given by equation1, where v is the meridional velocity, omega is the pressure vertical velocity and r is Earth's radius. It is convenient to define a stream function for mean meridional circulation that satisfies the zonally averaged continuity equation. The multiplicative constant 2*pi*r/g has been introduced here to keep the units of psi as kilograms per second. In order to estimate Polar Stream Function or psi the following Scheme is followed: -

$$1/\mathrm{r} * (\mathrm{d(v)/d(phi)}) + \mathrm{d}(omega)/\mathrm{d(p)} - [\mathrm{tan(omega)/r}] * \mathrm{v} = 0 \qquad (4.1)$$

$$1/\mathrm{r} * (\mathrm{d(v)} * \mathrm{cos(omega)})/\mathrm{d(phi)} + \mathrm{d(omega)/d(phi)} = 0 \qquad (4.2)$$

$$\mathrm{d(psi)/d(p)} = \mathrm{v} * 2 * \mathrm{pi} * \mathrm{r} * \mathrm{cos(phi)} \qquad (4.3)$$

$$1/\mathrm{r} * \mathrm{d(psi)/d(psi)} = -[\mathrm{omega} * 2 * \mathrm{pi} * \mathrm{r} * \mathrm{cos(phi)}]/[\mathrm{g}] \qquad (4.4)$$

i. Firstly, a long record of monthly wind observations, the mean zonally averaged meridional wind is calculated as a function of latitude and pressure.

ii. Next the vertical mean, zonally averaged vertical velocity is calculated by solving the continuity equation.

iii. Finally, the mean zonally averaged stream function is calculated by solving a system of below mentioned equations.

iv. Stream function is a convenient way of looking at circulation because it simultaneously represents both the meridional and vertical circulations

v. These stream functions are usually graphically represented and studied as isopleths.

vi. These isopleths are a representative of the Hadley Cell, the Polar cell and the Ferrel Cell which are the cells of mean meridional circulation.

4.2 Estimation of Wind Data Using Polar Stream Function Values After Applying Dyne's Compensation

The net mass convergence into a column of air (from the surface to the tropopause) is generally much smaller than the convergence at any particular level. This is because convergence at one level tends to be offset by divergence at another. This is known as *Dyne's compensation.* For any model such as NCEP/NCAR reanalysis dataset, the omega values generated are dependent on the vertical placement of pressure levels. For the purpose of this project, pressure levels different to those used in the NCEP/NCAR dataset have been used, hence, in attempt to compensate for the loss in accuracy and prediction., Dyne's compensation is applied, furthermore you are imposing Dyne's compensation, which NCEP/NCAR does not. Because of these differences V and Omega need to be consistent for your pressure levels.

i. The V-wind data directly obtained from reanalysis dataset cannot be as it is without any processing. Hence, it recalculated through iteration using the values obtained for the polar stream function in the previous step.
ii. This process involves applying Dyne's compensation.
iii. Hence, the value of 'V' which is obtained in this step is much more accurate than the reanalysis values and can be subjected to further computation.
iv. The equation used for calculating 'V' wind values using Polar Stream Function are: -

$$\mathrm{V} = -\left[\mathrm{g} - \{2 * \mathrm{pi} * \mathrm{a} * \cos(\mathrm{phi})\}\right] * \frac{\mathrm{d(psi)}}{\mathrm{dp}} \qquad (4.5)$$

4.3 Estimation of Air Density Using Temperature

i. Air density of values of the stratospheric ozone layer are computed using the NCEP/NCAR temperature dataset, the geographical location of a given points, which means its spatial resolution.

ii. The temperature dataset follows the same format as that of V-wind dataset.

iii. Air density is calculated.

iv. Using these values vertical wind velocity ‘w’ is calculated.

4.4 Estimation of Polar Cell

i. To estimate the value of polar cell, first the V-wind velocity and ‘W’ vertical wind velocity is both subjected to double averaging, *i.e.* averaged over both pressure and latitude.
ii. Now, for each year from 1974-2015 these values are summed up separately.
iii. Next the squares of these sums are calculated.
iv. These squares are again, summed up their square root is calculated.
v. This gives the value of polar cell.
vi. However, the value obtained at this step is not very precise.

4.5 Estimation of Climatological Fields

Climatological fields considered for this studied are polar jet stream and the subtropical jet stream, the 50 hPa level air temperatures and the convergence of flux of temperature at that level.

1. U-wind values at 300 hPa Level
2. Polar Jet & Subtropical Jet at 50 hPa Level.
3. Subtropical Rainfall
4. Temperature Values

4.5.1 U-Wind Values at 300 HPa Level

The procedure for U-wind climatology generation is as below: -

i. Once the major epochs of ozone depletion had been identified, those years were taken as the major point of consideration.
ii. These epochs occurred in the years were 1998, 2000, 2002, and 2006.
iii. These were the years of maximum recorded ozone lows.

iv. Now in an attempt to find out the influence of these values, months before and after ozone depletion period were considered.
v. U-wind values from NCEP/NCAR Reanalysis Dataset were taken for the months March to August for the years 1998, 2000, 2002 and 2006.
vi. The files containing these values were concatenated so that for all the years the U-wind values were stacked one after the other.
vii. Once the data had been arranged in this form, the U-wind values were averaged over time. These values were called as before values.
viii. Using visualization tools, isopleths of these values were prepared for a domain of 60 degree South to 60 degree North at 300 hPa level.
ix. Now, for the same years, U-wind values for the months, October to March were taken from NCEP/NCAR dataset at 300 hPa level. Such as the first data file contained U-wind values from 1998 October to 1999 March, the second data file contained U-wind values from 2000 October to 2001 March, the third data file contained U-wind values from 2002 October to 2003 March and the fourth file contained U-wind values from 2006 October to 2007 March.
x. Now, these values were processed in the same manner as before values *i.e* averaged over longitude, concatenated and then averaged over time and visualized using visualization tools. The files were called as after files. The final step in this process was to estimate the difference values. Once the after and before values had been averaged over longitude, concatenated, stacked and averaged over time, a difference in their values was calculated.
xi. These were again visualized.

4.5.2 Polar Jet and Subtropical Jet Values at 50 HPa Level

The procedure for Polar Jet and Subtropical Jet is as below: -

i. Once the major epochs of ozone depletion had been identified, those years were taken as the major point of consideration.
ii. These epochs occurred in the years were 1998,2000,2002 and 2006.
iii. These were the years of maximum recorded ozone lows.
iv. Now in an attempt to find out the influence of these values, months before and after ozone depletion period were considered.

v. U-wind values from NCEP/NCAR Reanalysis Dataset were taken for the months March to August for the years 1998, 2000, 2002 and 2006.
vi. The files containing these values were concatenated so that for all the years the U-wind values were stacked one after the other.
vii. Once the data had been arranged in this form, the U-wind values were averaged over time. These values were called as before values.
viii. Using visualization tools, isopleths of these values were prepared for a domain of 60 Degree South to 60 Degree North at 300 hPa level.
ix. Now, for the same years, U-wind values for the months, October to March were taken from NCEP/NCAR dataset at 300 hPa level. Such as the first data file contained U-wind values from 1998 October to 1999 March, the second data file contained U-wind values from 2000 October to 2001 March, the third data file contained U-wind values from 2002 October to 2003 March and the fourth file contained U-wind values from 2006 October to 2007 March.
x. Now, these values were processed in the same manner as before values *i.e* averaged over longitude, concatenated and then averaged over time and visualized using visualization tools. The files were called as after files.
xi. The final step in this process was to estimate the difference values. Once the after and before values had been averaged over longitude, concatenated, stacked and averaged over time, a difference in their values was calculated.
xii. These were again visualized.

4.5.3 Subtropical Rainfall Values

The procedure for Subtropical Rainfall climatology generation is as below: -

i. Once the major epochs of ozone depletion had been identified, those years were taken as the major point of consideration.
ii. These epochs occurred in the years were 1998,2000,2002 and 2006.
iii. These were the years of maximum recorded ozone lows.
iv. Now in an attempt to find out the influence of these values, months before and after ozone depletion period were considered.
v. Precipitation values from TRMM 3B43 Dataset were taken for the months March to August for the years 1998, 2000, 2002 and 2006.

vi. The files containing these values were concatenated so that for all the years the precipitation values were stacked one after the other.

vii. Once the data had been arranged in this form, the U-wind values were averaged over time. These values were called as before values.

viii. Using visualization tools, isopleths of these values were prepared.

ix. Now, for the same years, U-wind values for the months, October to March were taken from TRMM 3B43 dataset at 300 hPa level. Such as the first data file contained precipitation values from 1998 October to 1999 March, the second data file contained precipitation values from 2000 October to 2001 March, the third data file contained precipitation values from 2002 October to 2003 March and the fourth file contained precipitation values from 2006 October to 2007 March.

x. Now, these values were processed in the same manner as before values *i.e* averaged over longitude, concatenated and then averaged over time and visualized using visualization tools. The files were called as after files.

xi. The final step in this process was to estimate the difference values. Once the after and before values had been averaged over longitude, concatenated, stacked and averaged over time, a difference in their values was calculated.

xii. These were again visualized.

4.5.4 Temperature Values

The procedure for temperature climatology generation is as below: -

i. Once the major epochs of ozone depletion had been identified, those years were taken as the major point of consideration.

ii. These epochs occurred in the years were 1998,2000,2002 and 2006.

iii. These were the years of maximum recorded ozone lows.

iv. Now in an attempt to find out the influence of these values, months before and after ozone depletion period were considered.

v. Temperature Values from NCEP/NCAR Reanalysis Dataset were taken for the months March to August for the years 1998, 2000, 2002 and 2006.

vi. The files containing these values were concatenated so that for all the years the precipitation values were stacked one after the other.

vii. Once the data had been arranged in this form, the Temperature values were averaged over time. These values were called as before values.

viii. Using visualization tools, isopleths of these values were prepared.

ix. Now, for the same years, Temperature values for the months, October to March were taken from NCEP/NCAR Reanalysis Dataset. Such as the first data file contained precipitation values from 1998 October to 1999 March, the second data file contained precipitation values from 2000 October to 2001 March, the third data file contained precipitation values from 2002 October to 2003 March and the fourth file contained precipitation values from 2006 October to 2007 March.

x. Now, these values were processed in the same manner as before values *i.e* averaged over longitude, concatenated and then averaged over time and visualized using visualization tools. The files were called as after files.

xi. The final step in this process was to estimate the difference values. Once the after and before values had been averaged over longitude, concatenated, stacked and averaged over time, a difference in their values was calculated.

xii. These were again visualized.

CHAPTER 5 - RESULTS & DISCUSSIONS

A general analysis of monthly climatologies for all 42 years was carried out to understand the circulation pattern of the study area. The climatologies prepared were as depicted below:

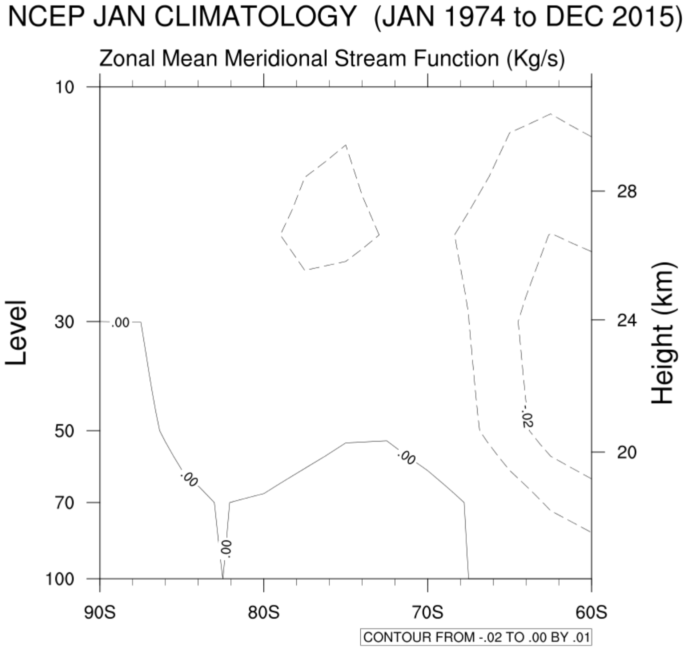


Figure 5.1.1: Jan Climatology

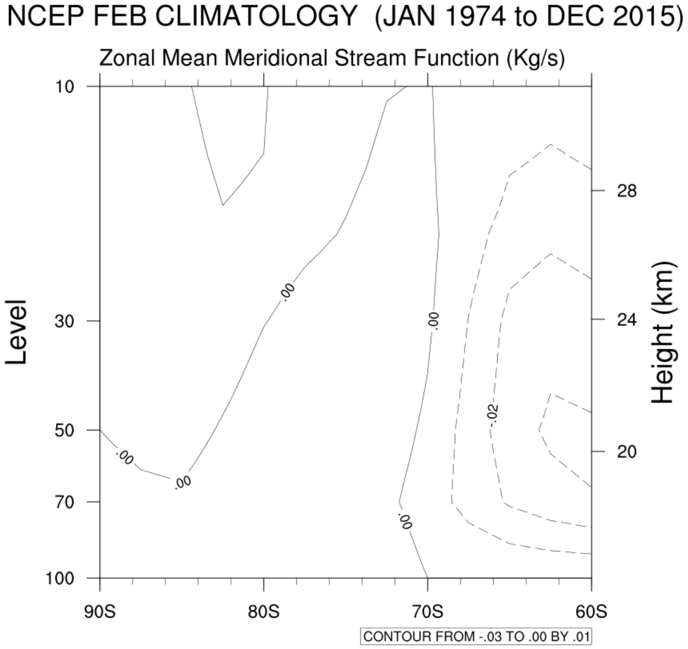


Figure 5.1.2: Feb Climatology

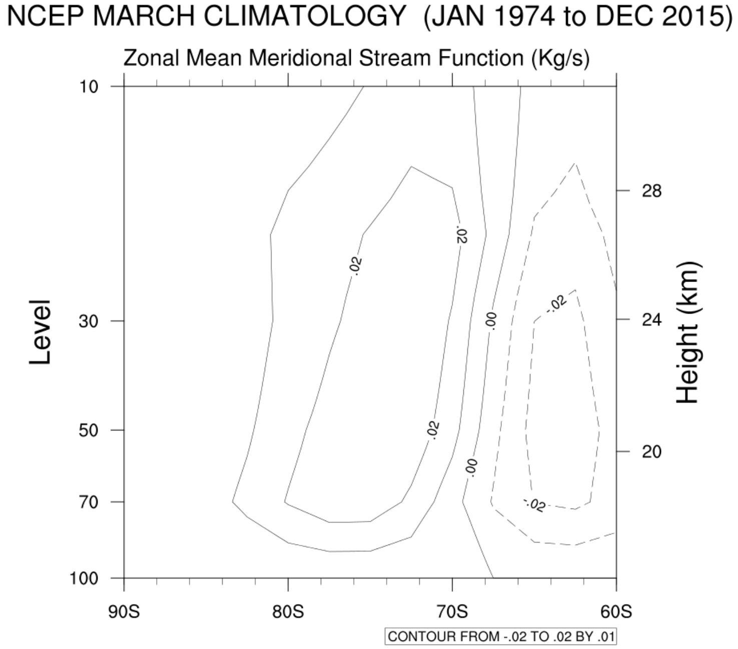


Figure 5.13 March Climatology

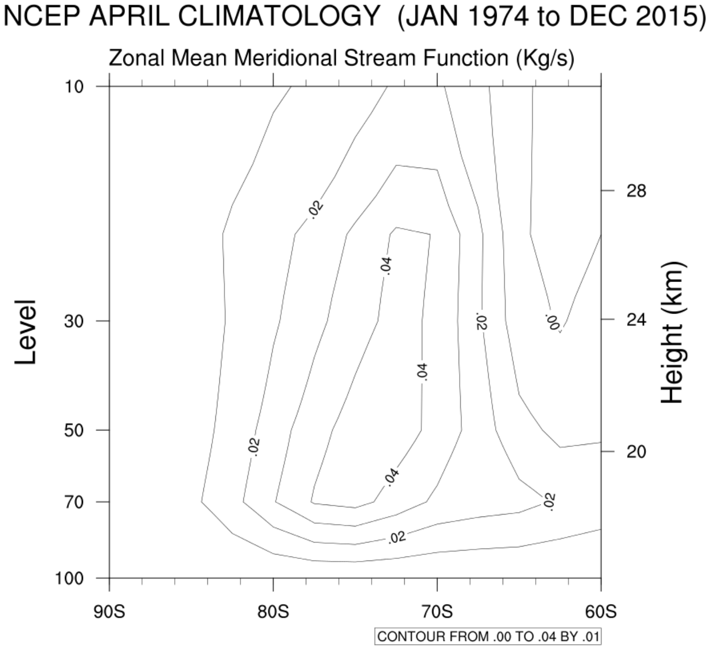


Figure 5.1.4: April Climatology

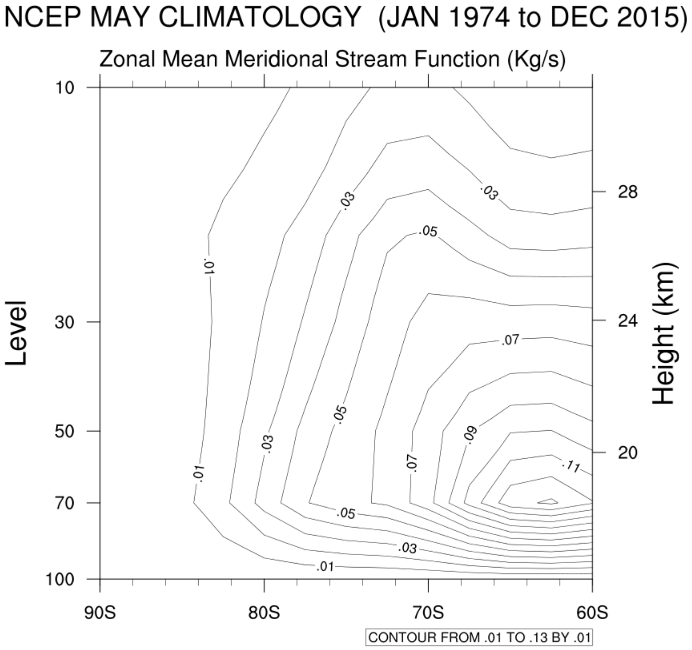


Figure 5.1.5: May Climatology

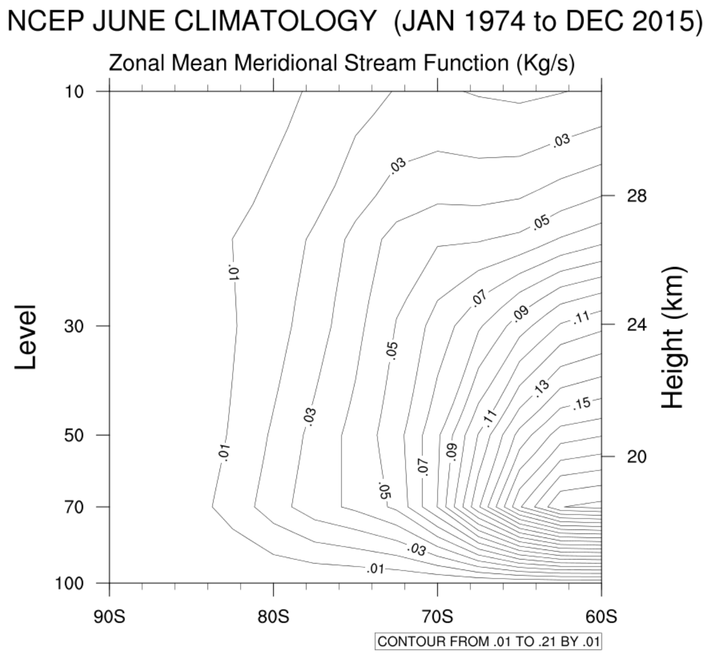


Figure 5.1.6 June Climatology

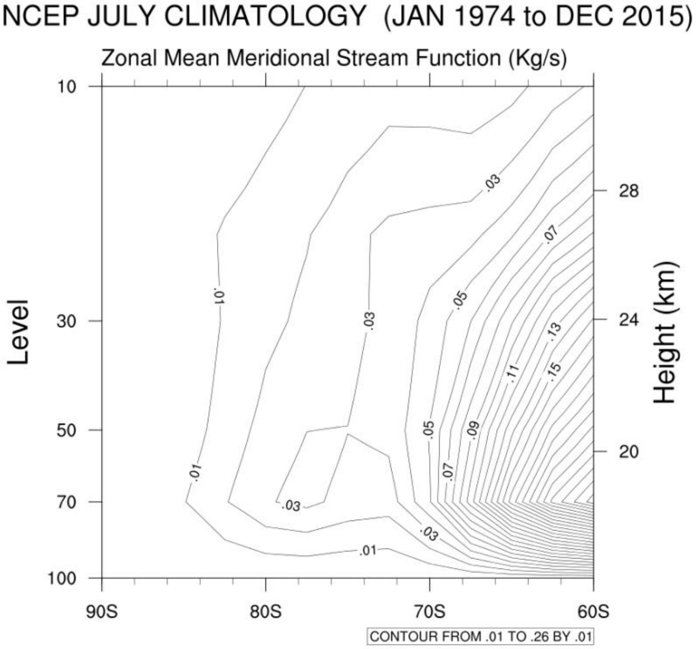


Figure 5.1.7: July Climatology

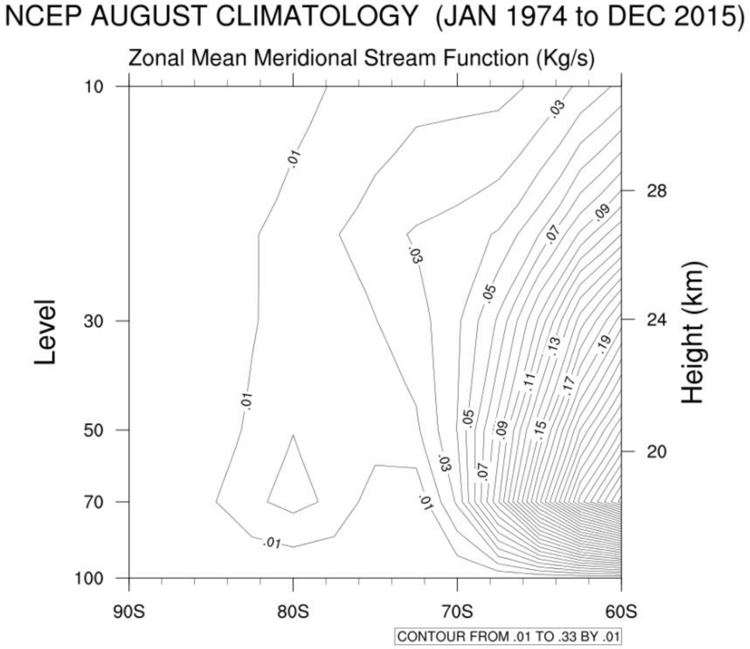
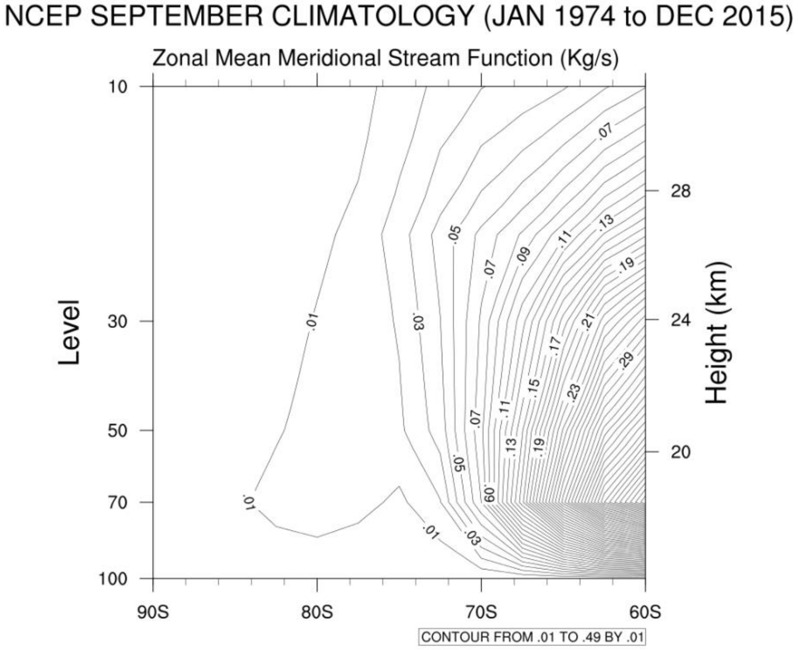
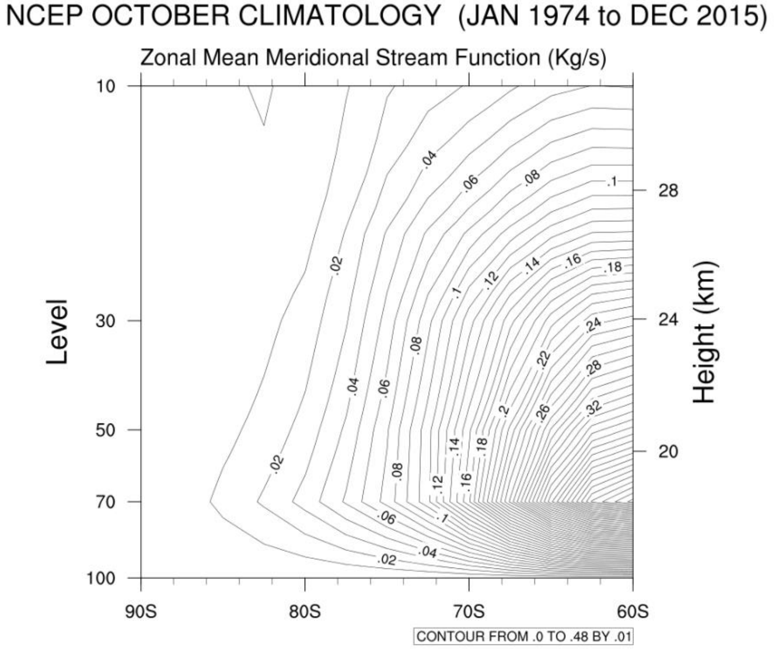


Figure 5.1.8: August Climatology


NCEP SEPTEMBER CLIMATOLOGY (JAN 1974 to DEC 2015)

Zonal Mean Meridional Stream Function (Kg/s)

Figure 5.1.10: September Climatology


NCEP OCTOBER CLIMATOLOGY (JAN 1974 to DEC 2015)

Zonal Mean Meridional Stream Function (Kg/s)

Figure 5.1.11: October Climatology

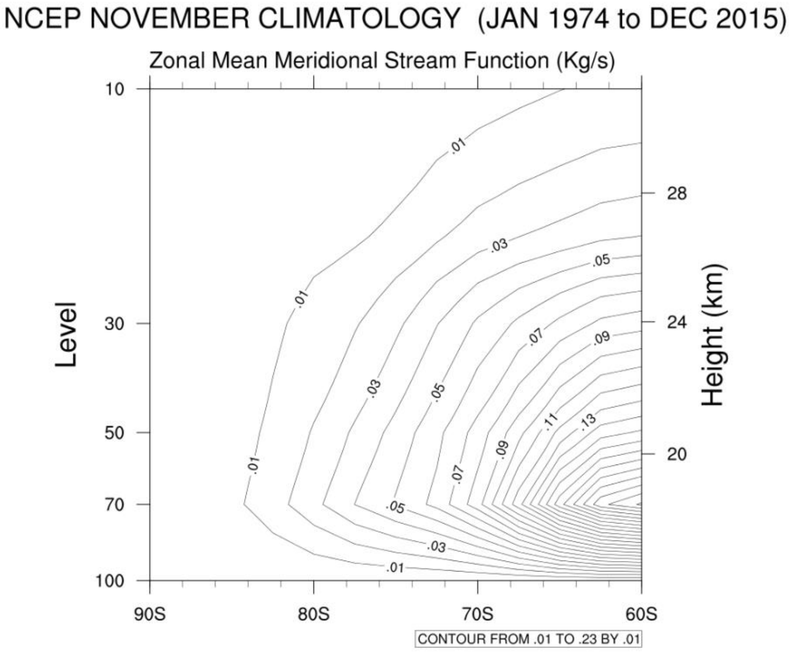


Figure 5.1.12: November Climatology

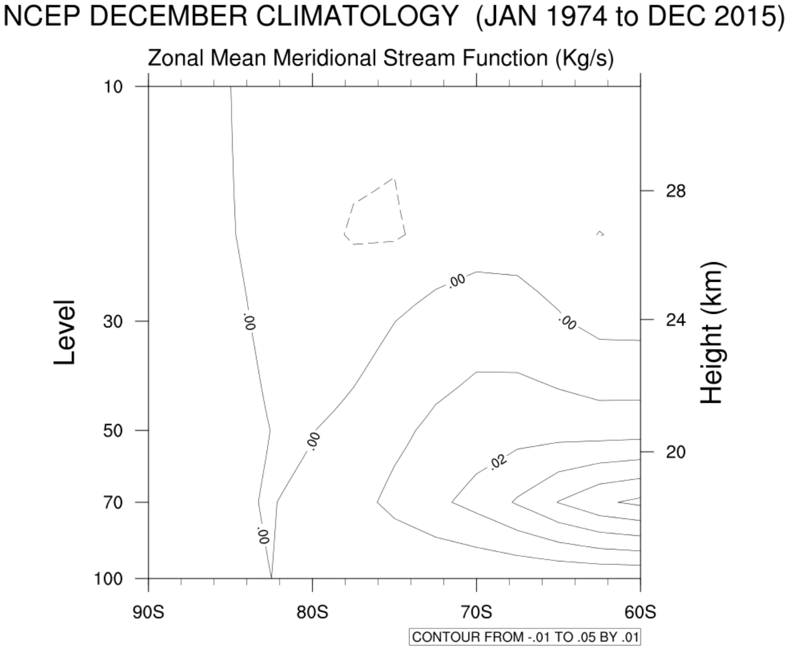


Figure 5.1.13: December Climatology

5.1 Polar Stratospheric Cell Formation

The construction of the stream function describing the stratospheric vertical circulations was discussed earlier in section. Those polar stratospheric cells were computed for the entire 492 months. Composite structures of these cells were also constructed before and after the formation of the most intense ozone hole during these 492 months. Those were noted during the months of September of 1994, 1998, 2000 and 2006. For these four years we made 6 months prior to and six months after these intense ozone hole months. Those averaged values of the stream function are displayed in figures. The prominent upward and downward lobes are marked in these two illustrations.

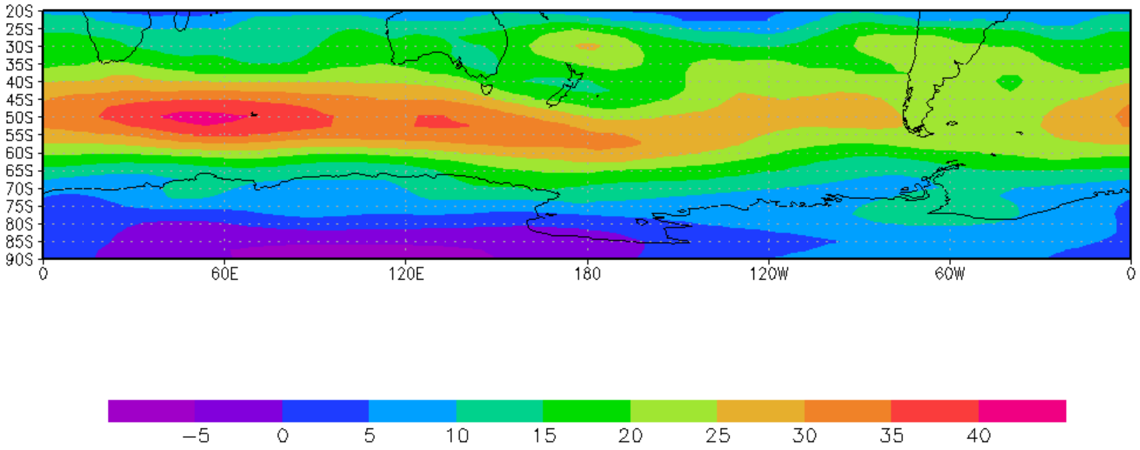


Figure 5.1.14

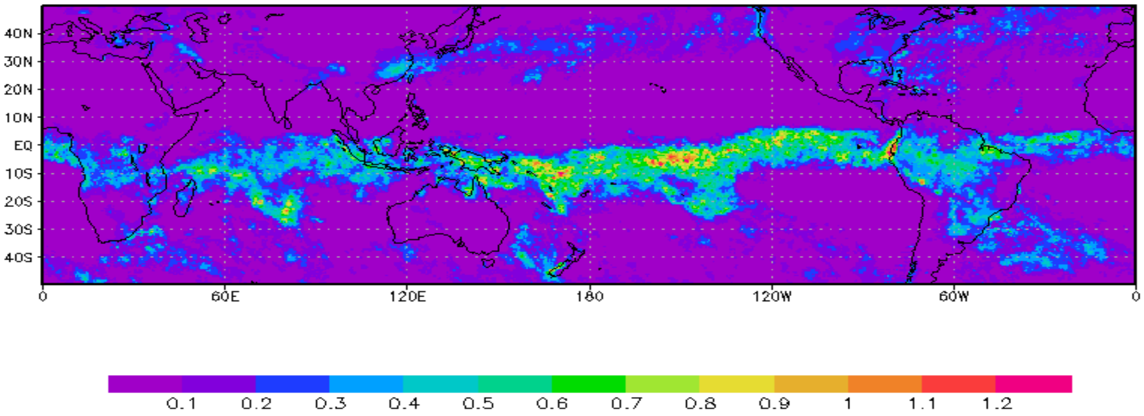


Figure 5.1.15

5.2 Polar Jet

In order to map the impact of polar jet on ozone depletion or formation for the given study, average climatic condition for years 1998, 2000, 2002 and 2006, which were the major epochs of maximum ozone depletion were studied. Within these years the climatology six months before and six months after the period of ozone hole which for period of six months from March to August before the ozone hole formation, and six months from October to March of the next year was analyzed. It was observed that Polar Jet tend to shift southward along the latitude of Tasmania. This result was obtained by taking the monthly 300 hPa level winds for four epochs of ozone hole intensification as a frame of reference.

Another climatological field that was studied through this mechanism was subtropical polar jet. The maximum wind of the subtropical jet (that is different from the Polar Jet) was found to be located near 30 degrees South latitude and at 200 hPa level. Even at 300 hPa level we can always see a signature of the subtropical jet. During the period under observation, the subtropical jet showed a distinct southward shift as well, a result which was not noted by *Meehl et. al.* (*J Climate, 2012*). This is a new finding of this study. If one considered 6 months before the formation of the most intense ozone hole to a similar period after it, then it was observed that the subtropical jet strengthened and was located to 25 to 30 S latitudes over Australia, south America and the southern Atlantic Ocean. The polar jet and the subtropical jet were found to merge over the southern Atlantic into a broad belt of westerlies six months before the most intense ozone hole strengthening the intensity of subtropical jet. The difference in intensity was found to be much as 20 meters per second. The implication of this results is that there was found to be a southward shift of the tropical Hadley cell, during the intensification of the ozone hole, that generated the subtropical jet through the angular momentum principle, *Krishnamurti* (1960).

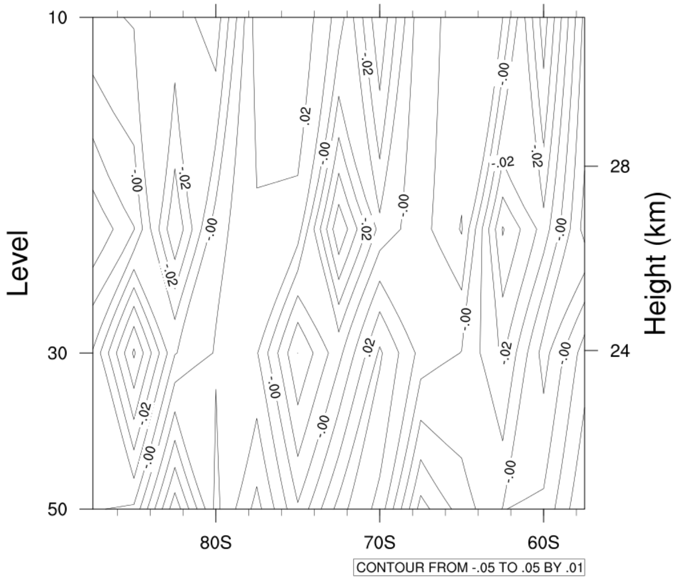


Figure 5.1.17

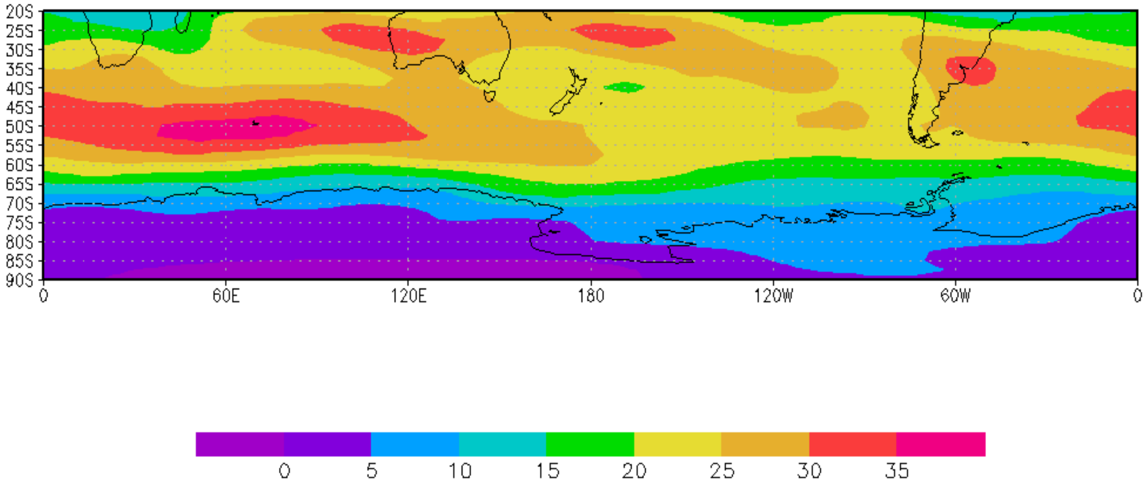
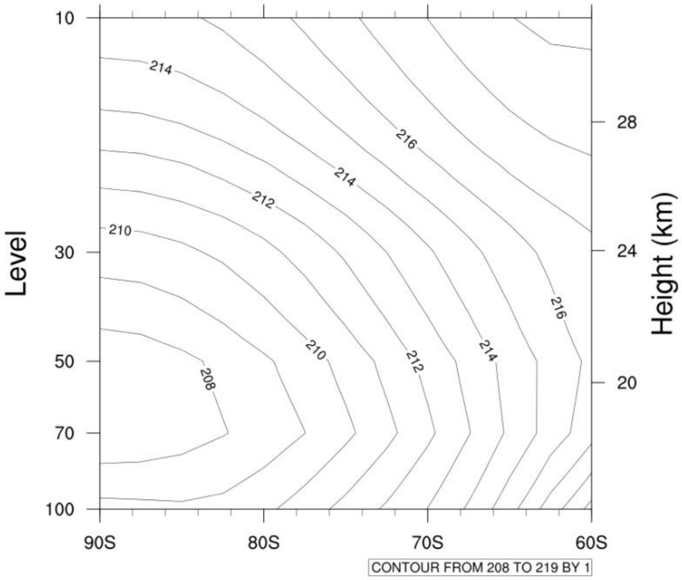


Figure 5.1.18



Level
Height (km)
10
30
50
70
100
28
24
20
90S
80S
70S
60S
214
216
214
212
210
210
212
214
216
208
CONTOUR FROM 208 TO 219 BY 1

Figure 5.1.20

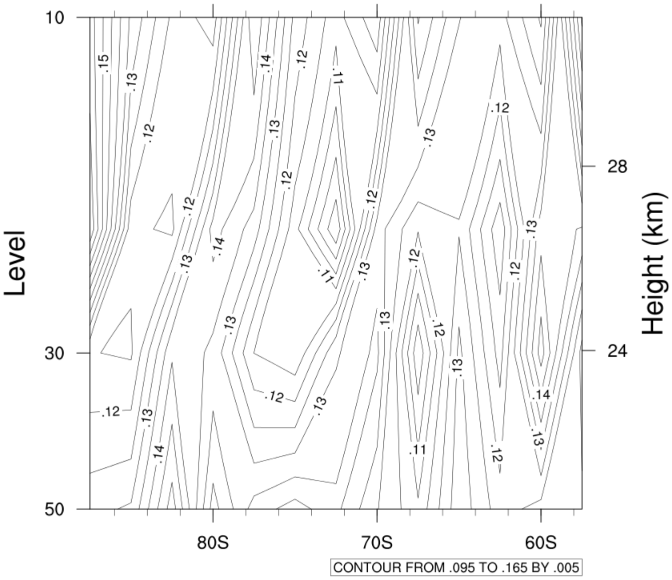

Level
Height (km)
10
30
50
28
24
80S
70S
60S
CONTOUR FROM .095 TO .165 BY .005
.15
.13
.12
.14
.11
.12
.13

Figure 5.1.21

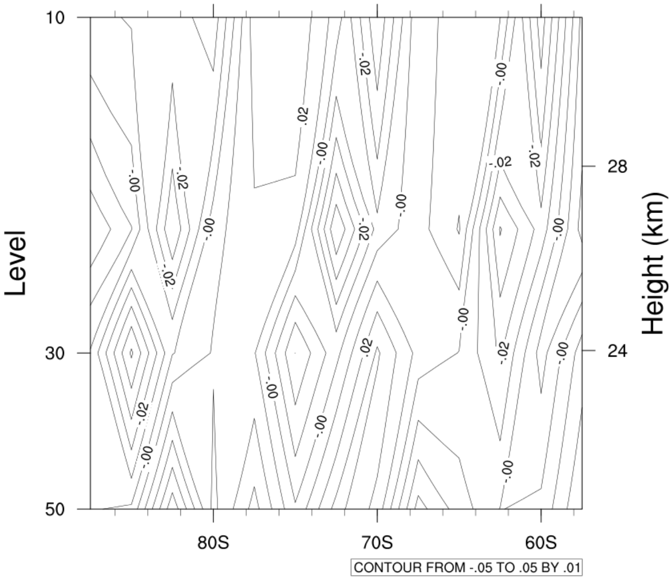

10
30
50
Level
80S
70S
60S
28
24
Height (km)
CONTOUR FROM -.05 TO .05 BY .01
.00
.02
-.02

Figure 5.1.22

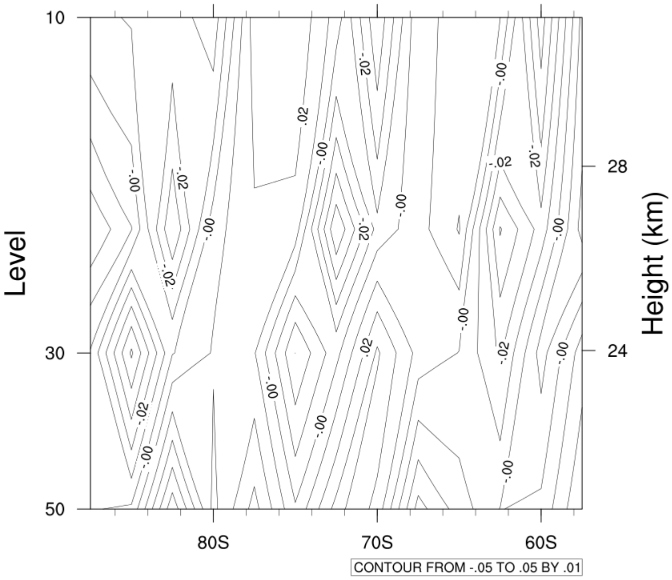

10
30
50
Level
80S
70S
60S
28
24
Height (km)
CONTOUR FROM -.05 TO .05 BY .01
.00
.02
-.02

Figure 5.1.23

5.4 Subtropical Rainfall

While previous studies have taken into account the influence of a variety climatological fields over ozone depletion and vice-versa, the impact of subtropical rainfall over ozone depletion is still understudied. This study has been able to find teleconnections in between subtropical rainfall and ozone depletion. Major events of spring associated ozone depletions over a duration of 41 years from 1975 to 2014 were taken into account here and their link with the subtropical rainfall pattern over ozone depletion was studied. Subtropical rainfall encompasses the rainfall in regions lying in between the Tropic of Cancer and the Tropic of Capricorn. This roughly spans the entire domain extending from equator to nearly 23.27 degree latitudes on either side of the equator. For the purpose of this study TRMM data was used. The data was available for entire duration of study. This was in fact, the off-equatorial rainfall maximum of the oceanic Inter Tropical Convergence Zone, especially over the Pacific Ocean. This precipitation data was available in millimeters per day. The Tropical Rainfall Measuring Mission (TRMM) is a joint mission between NASA and the Japan Aerospace Exploration (JAXA) Agency to study rainfall for weather and climate research. The satellite with a mission to collect global tropical rainfall and lightning data was launched in late November 1997 and continued up till April 15, 2015. Although the equipments were designed to work for a period of 3years however they continued to work for a long period of 17 years. The satellite contained 5 instruments in all, one was a 3-sensor rainfall suite (PR, TMI, VIRS) and 2 related instruments (LIS and CERES). The data

obtained through TRMM has been popularly used in projects related to tropical cyclone structure and evolution, convective system properties, lightning-storm relationships, climate and weather modeling, and human impacts on rainfall. Apart from the above mentioned research applications it has also been found to have operational applications such as flood and drought monitoring and weather forecasting. Periods under observation noted a latitudinal shift in the subtropical rains during the presence of the ozone hole. Figures show the two scenarios of this latitudinal shift towards the higher latitudes of the southern hemisphere.

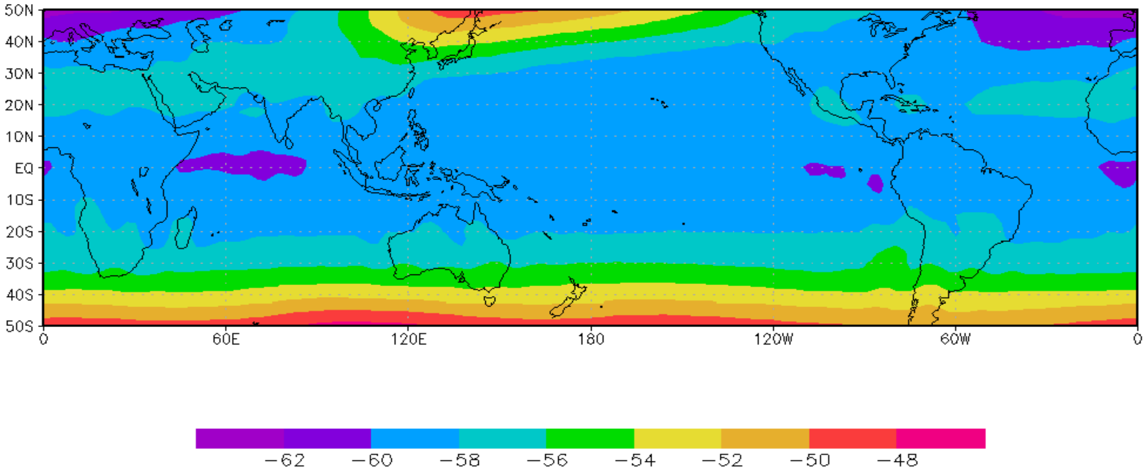
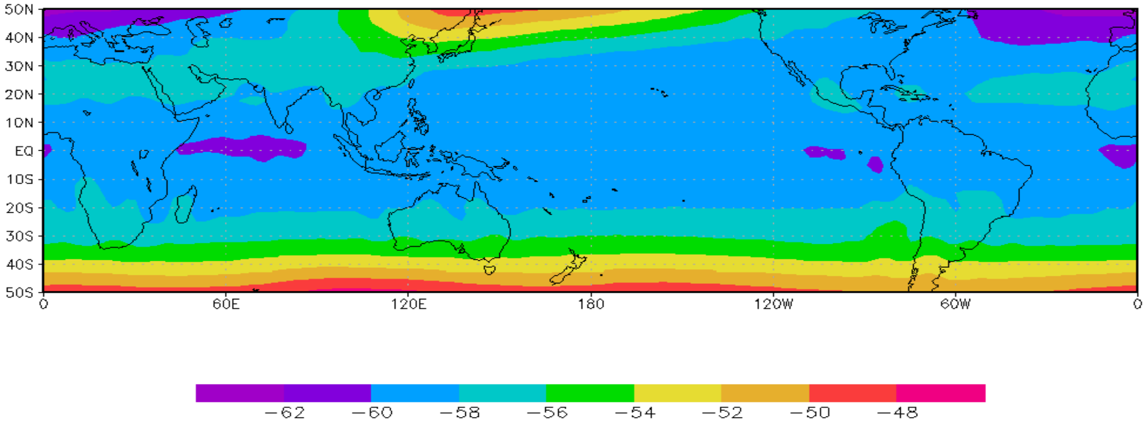

50N
40N
30N
20N
10N
EQ
10S
20S
30S
40S
50S
0
60E
120E
180
120W
60W
0
−62
−60
−58
−56
−54
−52
−50
−48

Figure 5.1.24


50N
40N
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EQ
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60W
0
−62
−60
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−54
−52
−50
−48

Figure 5.1.25

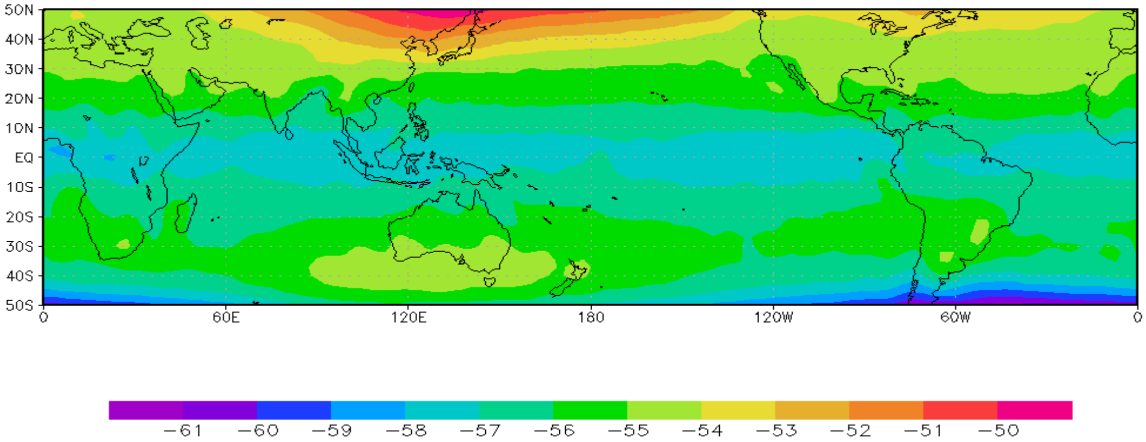


Figure 5.1.26

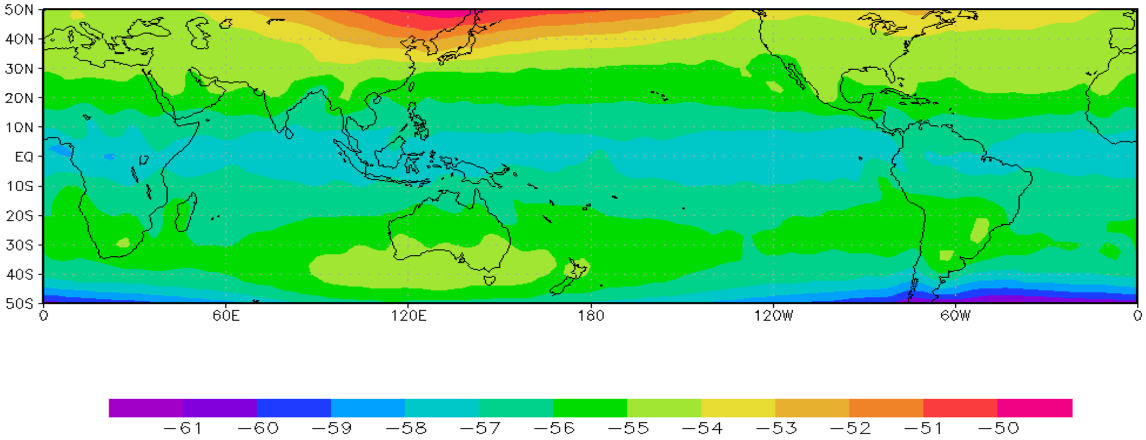
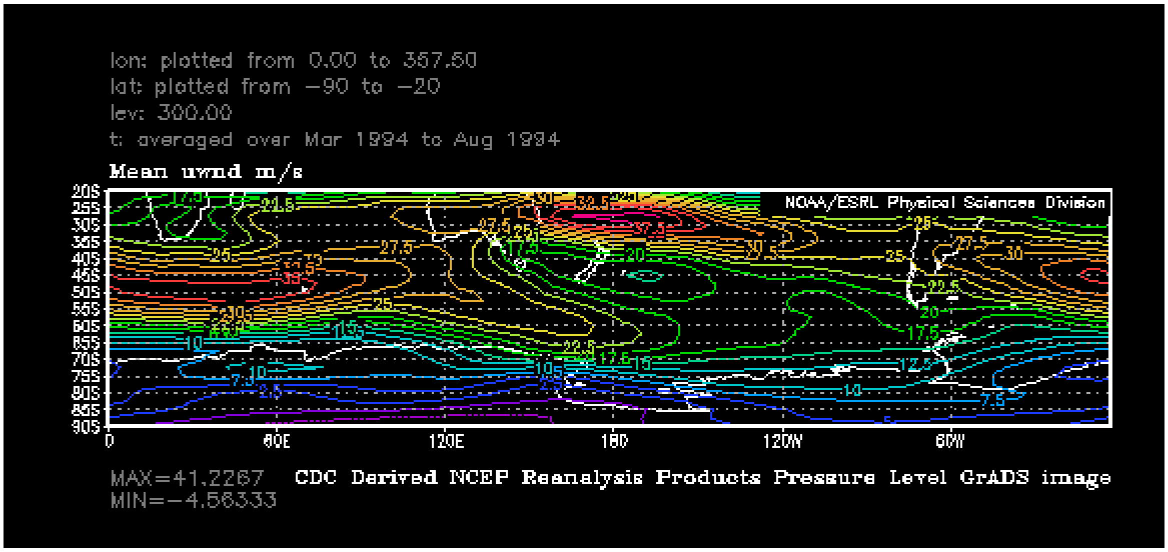


Figure 5.1.27



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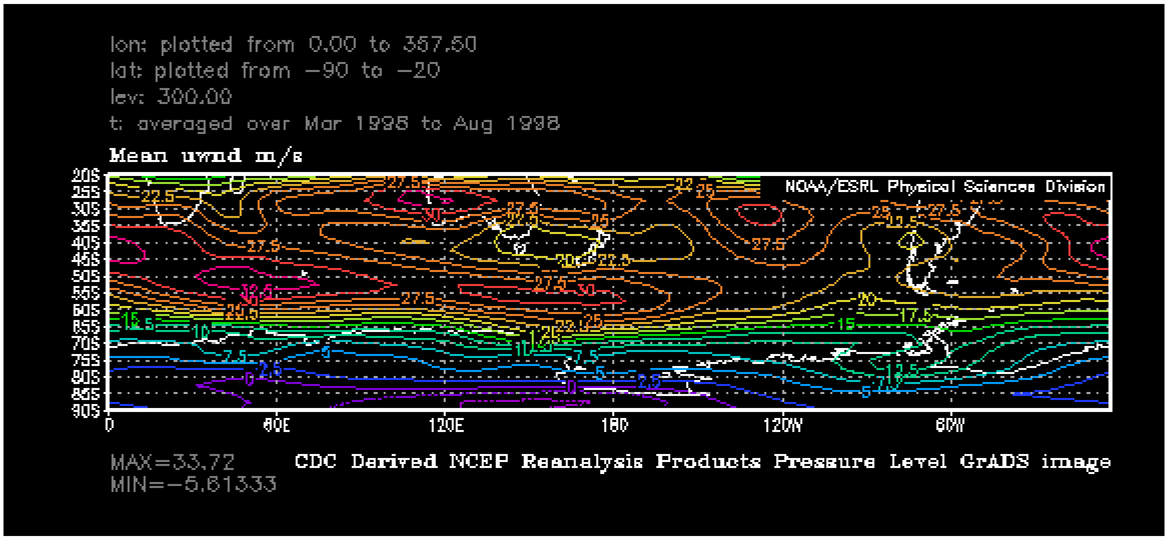

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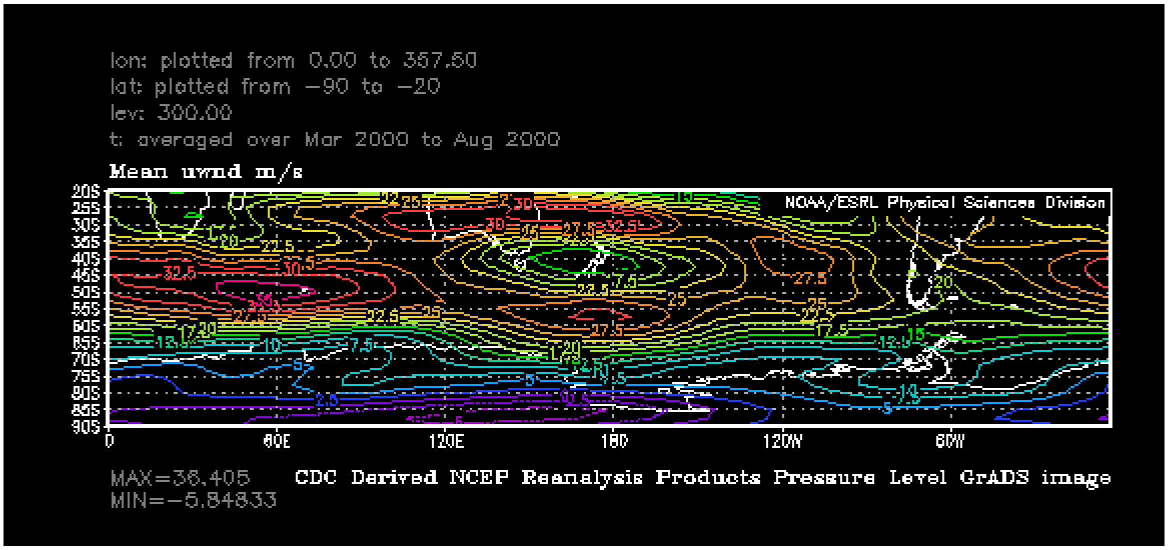
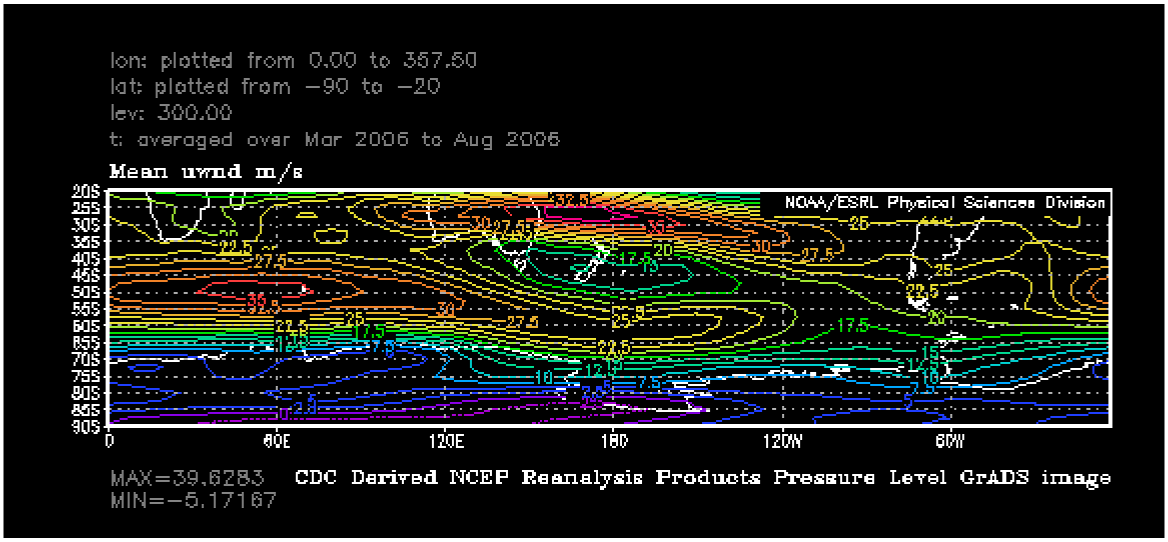


Figure 5.1.30



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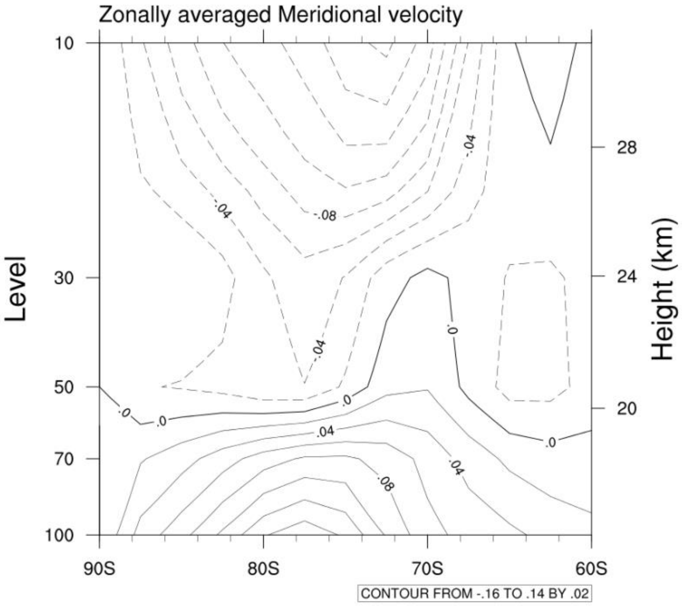

Zonally averaged Meridional velocity
Level
Height (km)
10
30
50
70
100
28
24
20
90S
80S
70S
60S
-.04
-.08
-.04
-.04
.0
.0
.0
.0
.0
.0
.04
.04
.08
CONTOUR FROM -.16 TO .14 BY .02

Figure 5.1.32

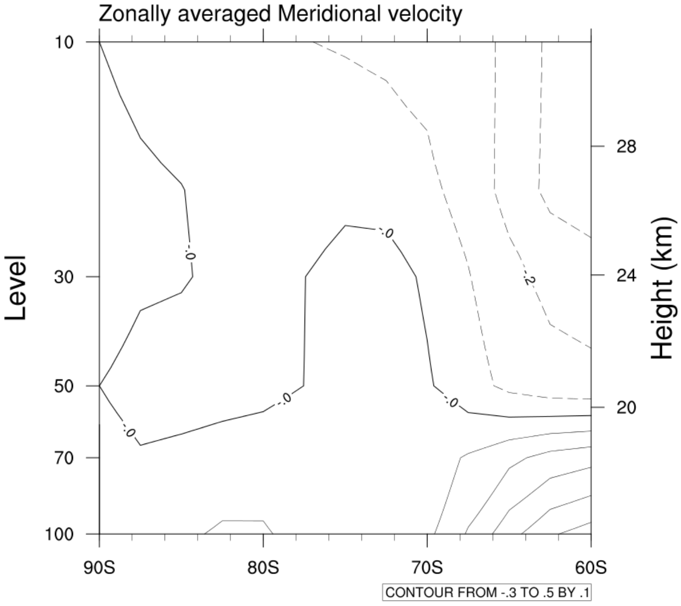

Zonally averaged Meridional velocity
Level
Height (km)
10
30
50
70
100
28
24
20
90S
80S
70S
60S
.0
.0
.0
.0
.0
-.2
CONTOUR FROM -.3 TO .5 BY .1

Figure 5.1.33

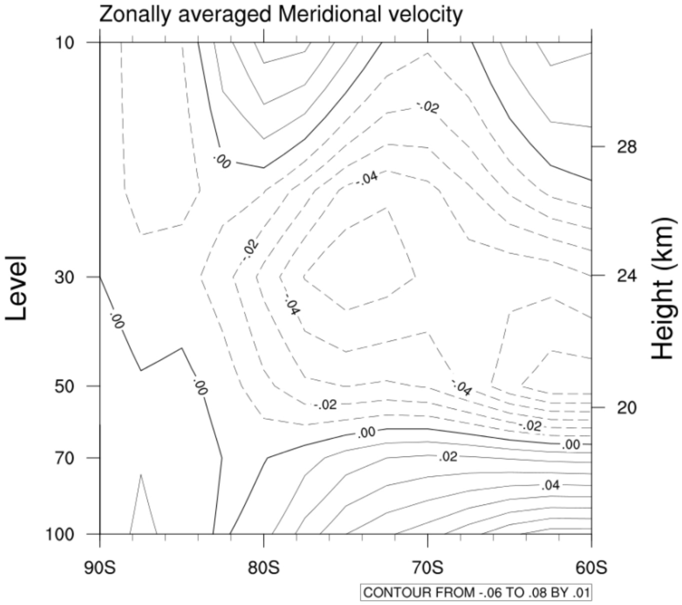


Figure 5.1.34

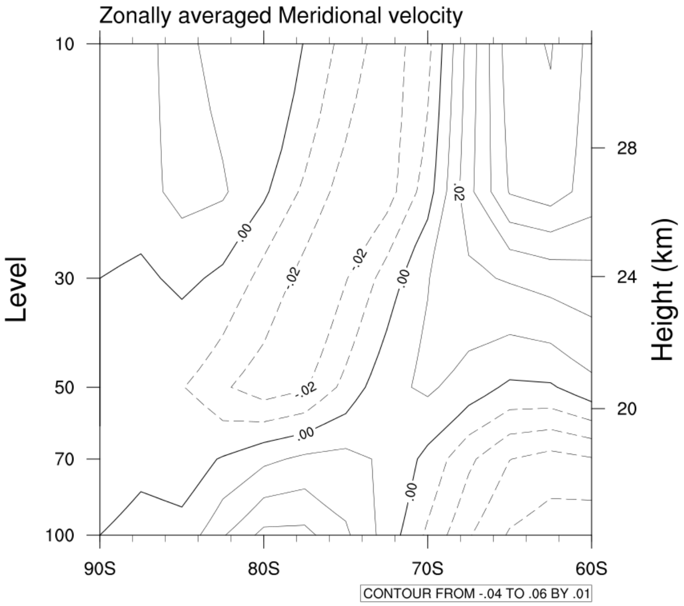
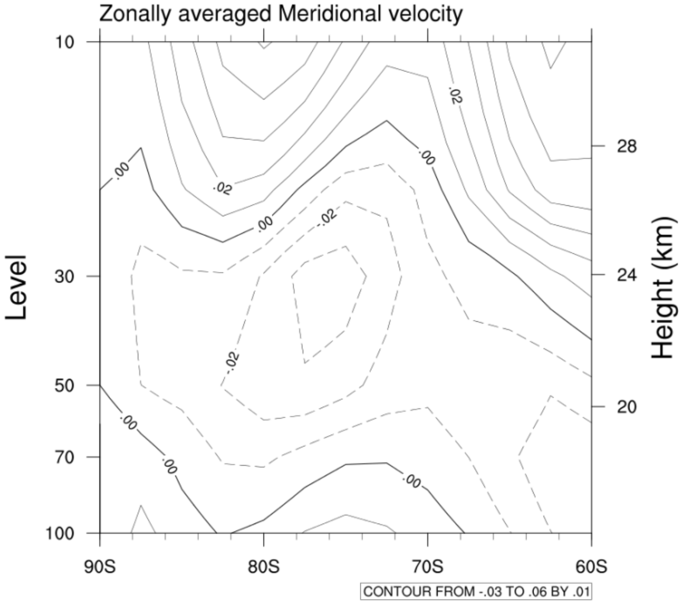

Zonally averaged Meridional velocity
Level
10
30
50
70
100
Height (km)
28
24
20
90S
80S
70S
60S
.00
-.02
-.02
.00
-.02
.00
.00
.02
CONTOUR FROM -.04 TO .06 BY .01

Figure 5.1.35


Zonally averaged Meridional velocity
Level
10
30
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90S
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70S
60S
Height (km)
28
24
20
.02
.00
.02
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.02
.00
.02
.00
.00
.00
.00
CONTOUR FROM -.03 TO .06 BY .01

Figure 5.1.36

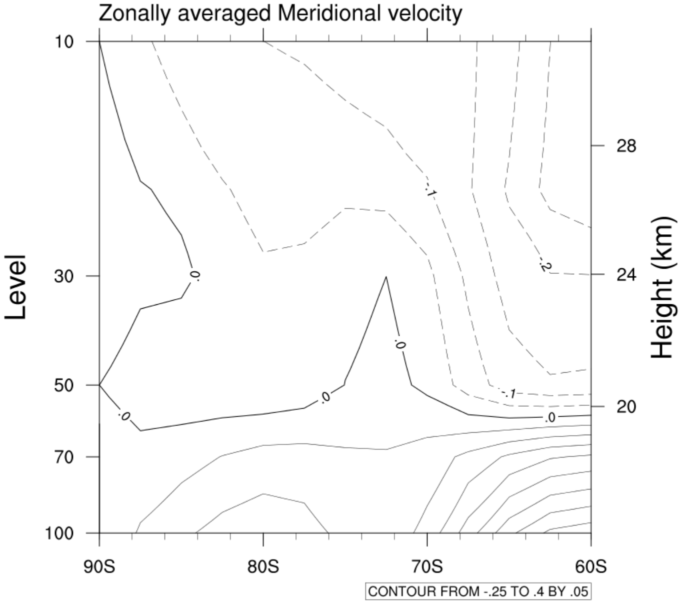

Zonally averaged Meridional velocity
Level
10
30
50
70
100
90S
80S
70S
60S
Height (km)
28
24
20
-.1
-.2
.0
.0
-.1
.0
.0
.0
.0
CONTOUR FROM -.25 TO .4 BY .05

Figure 5.1.37

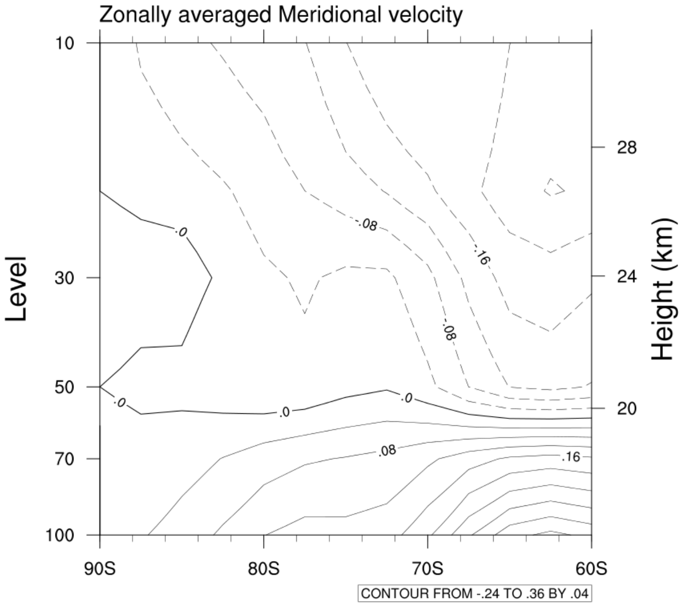
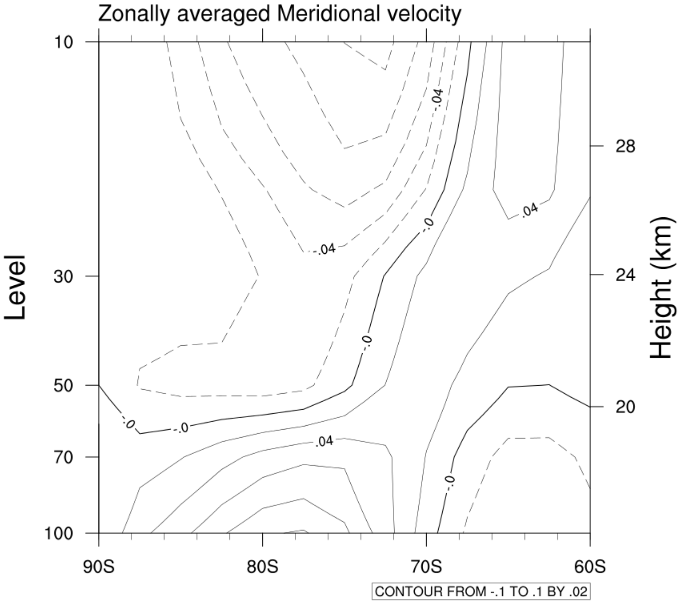


Figure 5.1.38


Zonally averaged Meridional velocity
Level
Height (km)
10
30
50
70
100
28
24
20
90S
80S
70S
60S
-.04
.04
-.0
.0
CONTOUR FROM -.1 TO .1 BY .02

CHAPTER 6 – CONCLUSIONS

The impact of climatological fields on ozone was studied. In case of stratospheric polar cell studies, what is noted here is an upward extension of strong lobe of upward motion near 85 degrees south as we go from the before period towards the after period. This upward lobe is part of an elongated vertical circulation cell that connects the lower stratosphere with the upper stratosphere and is located south poleward of 80 Degrees south latitude. This strongly suggests that in the region of formation of the ozone hole a prominent lobe of upward motion forms that carves its way upward.

Polar jet impact studies show in general that most of the principle activities of the general circulation show a southward shift, such as the polar jet, the Ferrell cell etc. In this context it should be noted that the altitude of maximum winds for the polar jet was found to be near 300 hPa surface whereas in case of subtropical jet, it generally has its maximum intensity near 175 hPa over Australia and the Southern Atlantic Ocean. Since the mass of the atmosphere participating in the formation of the ozone hole in the stratosphere is very much smaller compared to these tropospheric features, it is safe to conclude that the ozone hole intensification is a consequence of these major changes in the troposphere. So if we see large changes going on before September in the troposphere we can anticipate a possible strengthening of the ozone hole. A more definitive study can only come from numerical modeling where forecasts from such antecedents should contribute to an intensification of the ozone hole. Such a modeling study should invoke also the effects of chlorofluorocarbons, to see its place among others, within this scenario.

Subtropical rainfall studies show that what this rainfall does is to provide a poleward shift of the tropospheric Hadley cell, which in turn sequentially impacts the Ferrel cell and the location of the polar jet of the southern hemisphere. Our data sets essentially confirm these same features on the shift of the subtropical rains.

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