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DYNAMICAL PROCESSES IN THE STRATOSPHERE AND UPPER TROPOSPHERE AND THEIR INFLUENCE ON THE DISTRIBUTION OF TRACE GASES IN THE POLAR ATMOSPHERE

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Abstract

Transport plays an important role in the distribution of long-lived gases such as ozone and water vapour in the atmosphere. Understanding of observed variability in these gases as well as prediction of the future changes depends therefore on our knowledge of the relevant atmospheric dynamics. This dissertation studies certain dynamical processes in the stratosphere and upper troposphere which influence the distribution of ozone and water vapour in the atmosphere.

The planetary waves that originate in the troposphere drive the stratospheric circulation. They influence both the meridional transport of substances as well as parameters of the polar vortices. In turn, temperatures inside the polar vortices influence abundance of the Polar Stratospheric Clouds (PSC) and therefore the chemical ozone destruction. Wave forcing of the stratospheric circulation is not uniform during winter. The November-December averaged stratospheric eddy heat flux shows a significant anticorrelation with the January-February averaged eddy heat flux in the midlatitude stratosphere and troposphere. These intraseasonal variations are attributable to the internal stratospheric vacillations. In the period 1979-2002, the wave forcing exhibited a negative trend which was confined to the second half of winter only. In the period 1958-2002, area, strength and longevity of the Arctic polar vortices do not exhibit significant long-term changes while the area with temperatures lower than the threshold temperature for PSC formation shows statistically significant increase. However, the Arctic vortex parameters show significant decadal changes which are mirrored in the ozone variability. Monthly ozone tendencies in the Northern Hemisphere show significant correlations ($|\mathbf{r}|=0.7$) with proxies of the stratospheric circulation. In the Antarctic, the springtime vortex in the lower stratosphere shows statistically significant trends in temperature, longevity and strength (but not in area) in the period 1979-2001.

Analysis of the ozone and water vapour vertical distributions in the Arctic UTLS shows that layering below and above the tropopause is often associated with poleward Rossby wave-breaking. These observations together with calculations of cross-tropopause fluxes emphasize the importance of poleward Rossby wave breaking for the stratosphere-troposphere exchange in the Arctic.

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Dynaamiset prosessit stratosfäärissä ja ylemmässä troposfäärissa ja niiden vaikutus hivenkaasujen leviämiseen napa-alueiden ilmakehässä

Tiivistelmä

Kuljetuksella on tärkeä rooli otsonin ja vesihöyryn leviämisessä ilmakehässä. Siksi näiden kaasujen havaitun vaihtelun ymmärtäminen ja tulevaisuuden muutoksen ennustaminen vaatii tietoa asiaan vaikuttavasta ilmakehän dynamiikasta. Tämä väitöskirja tutkii eräitä otsonin ja vesihöyryn kuljetukseen liittyviä dynaamisia prosesseja stratosfäärissä ja ylemmässä troposfäärissä.

Planetaariset aallot, jotka syntyvät troposfäärissä, säätelevät stratosfäärin meridionaalista kiertoliikettä, joka puolestaan kuljettaa sekä materiaa että lämpöä päiväntasaajalta korkeammille leveysasteille ja siten mm. vaikuttaa polaarivorteksin ominaisuuksiin. Vorteksin lämpötilat vaikuttavat vuorostaan polaaristratosfääripilvien runsauteen ja siten otsonikatoon. Aaltovaikutus stratosfäärin kiertoon vaihtelee talven aikana. Marras-joulukuun keskimääräinen stratosfäärin lämpövuo näyttää selkeitä merkkejä antikorrelaatiosta tammi-helmikuun keskimääräisen stratosfäärin ja troposfäärin lämpövuon kanssa. Nämä vuodenaikavaihtelut ovat johdettavissa sisäisesta stratosfäärin värähtelystä. Aaltoaktiivisuus tarkastelukauden 1979-2002 aikana näytti tammi-helmikuussa negatiivista trendiä. Arktisen vorteksin pinta-ala, voimakkuus, ja kesto jakson 1958-2002 aikana eivät näyttäneet selkeitä merkkejä pitkäaikaisesta muutoksesta. Sen sijaan pinta-ala, jolla lämpötila on riittävän kylmä polaaristratosfääripilvien muodostumiselle, kasvoi tarkastelujaksolla hieman. Arktisen polaarivorteksin pitkäaikaiset muutokset muistuttavat otsonikerroksen muutoksia. Kuukausittain tarkasteltuna otsonitendenssit pohjoisella pallonpuoliskolla korreloivat selkeästi ($|r|=0.7$) stratosfäärin kiertoa edustavien muuttujien kanssa. Antarktisen alueella kevätaikainen polaarivorteksi jakson 1979-2001 aikana näyttää selkeää laskevaa lämpötilatrendiä. Vorteksin voimakkuus ja kesto olivat kasvussa, mutta vorteksin pinta-alassa ei havaita merkittävää muutosta.

Otsonin ja vesihöyryn pystykuljetuksen analyysi Arktisessa ylätroposfäärissä ja alastratosfäärissä osoittaa että anomaaliset kerrokset tropopausin ala- ja yläpuolella usein liittyvät napasuuntaisen Rossby-aallon murtumiseen. Nämä havainnot sekä laskelmat kaksisuuntaisesta kuljetuksesta tropopaussin läpi osoittavat kuinka tärkeä merkitys napasuuntaisen Rossby-aallon murtumisella on stratosfääri-troposfäärivaihdossa arktisessa yläilmakehässä.

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Lisätietoja

This work is dedicated to Dasha

PREFACE

The work presented in this thesis has been carried out at the Finnish Meteorological Institute (FMI) in 2002-2007. I would like to thank my supervisor Prof. Esko Kyrö, who offered me the excellence opportunity to work at the FMI, for the patient guidance he provided throughout my studies and for his continuous encouragement.

During this work, discussions with my friend and colleague Dr. Grigory Nikulin were extremely helpful. I appreciate help of Dr. Alexander Lukyanov (CAO, Moscow) who let me use his trajectory model. I'm very grateful to Prof. Erkki Kyrölä and Dr. Juhanni Damski who read this manuscript carefully and provided useful comments. Prof. Hannu Savijärvi is acknowledged with great gratitude for making the defence of this dissertation possible at the University of Helsinki. Special thanks are due to Dr. Robin King who checked my writings and improved the language of this thesis substantially. I would also like to thank my first supervisor Dr. Mikhail Beloglazov (Polar Geophysical Institute, Russia) who helped me during my first steps in the atmospheric science.

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ABBREVIATIONS

AASE	Airborne Arctic Stratosphere Expedition
BD	Brewer-Dobson
CAT	Clear-Air Turbulence
CFC	Chlorofluorocarbon
ECMWF	European Centre for Medium-Range Weather Forecasts
ENSO	El Nino Southern Oscillation
EP	Eliassen-Palm
ERA	ECMWF Re-Analysis
HALOE	Halogen Occultation Experiment
HF	Heat Flux
JF	January-February
NCAR	National Center for Atmospheric Research
NCEP	National Centers for Environmental Prediction
ND	November-December
NH	Northern Hemisphere
PSC	Polar Stratospheric Cloud
PV	Potential Vorticity
PVU	Potential Vorticity Unit
QBO	Quasi-Biannual Oscillation
SAGE	Stratospheric Aerosol and Gas Experiment
SH	Southern Hemisphere
STE	Stratosphere-Troposphere Exchange
TEM	Transformed Eulerian Mean
UTLS	Upper Troposphere Lower Stratosphere
WMO	World Meteorological Organization

CONTENTS

LIST OF PUBLICATIONS

PAPER 1: Karpetchko, A., Kyro, E., and von der Gathen, P., 2003. Generation of layering in the upper arctic troposphere away from the jet stream. *Annales Geophysicae 21*, p.1653-1665

PAPER 2: Karpetchko, A. and Nikulin, G., 2004. Influence of early winter upward wave activity flux on midwinter circulation in the stratosphere and troposphere. *J. Climate, v.17*, *N22*, p. 4443-4452.

PAPER 3: Karpetchko, A., Kyrö, E. and Knudsen, B. M., 2005. Arctic and Antarctic polar vortices 1957–2002 as seen from the ERA-40 reanalyses. *J. Geophys. Res.*, 110, D21109, doi:10.1029/2005JD006113.

PAPER 4: Nikulin, G., and Karpechko, A., 2005. The mean meridional circulation and midlatitude ozone buildup, *Atmos. Chem. Phys., 5*, p. 3159-3172.

PAPER 5: Karpechko, A., Lukyanov, A., Kyrö, E., Khaikin, S., Korshunov, L., Kivi, R. and Vömel, H., 2007. The water vapour distribution in the Arctic lowermost stratosphere during the LAUTLOS campaign and related transport processes including stratosphere-troposphere exchange. *Atmos. Chem. Phys., 7,* p. 107-119.

In PAPERS 1, 3, and 5, A. Karpechko was responsible for the major part of the calculations as well as for writing the manuscripts. In PAPER 2, A. Karpechko was involved in designing the objectives, making the calculations, interpretation of the results, and writing the manuscript. In PAPER 4, A. Karpechko was responsible for the data preparation, was involved in interpretation of the results and contributed to writing the manuscript.

1. INTRODUCTION

After the discovery of a deep spring minimum in the seasonal behaviour of the Antarctic ozone amount, the so-called 'Antarctic ozone hole' (*Farman et al., 1985*), and after the revelation of negative trends in ozone in both hemispheres (*Stolarski et al., 1991*), a lot of attention has been given to the middle atmosphere. Ozone changes were first attributed to the release of chlorofluorocarbons (CFCs) into the atmosphere as a result of human activity. However, attempts to explain the observed magnitude and spatial structure of the trends by chemistry alone were not successful (*Jackman et al., 1996, Solomon et al 1996*). Though the importance of dynamical processes for ozone changes, both on seasonal and interannual timescales, was put forward soon after the ozone hole discovery (*e.g. Tung and Yang, 1988; McIntyre 1989*), it took about a decade before *Fusco and Salby* (*1999*) clearly showed that the ozone changes in the Northern Hemisphere (NH) are in fact strongly coupled to changes in the stratospheric meridional circulation. The main question raised by these studies was: which part of the ozone changes is explained by a variability in the dynamics and which part is caused by perturbation of the chemical composition of the atmosphere due to human impact? It was also realized that changes in ozone can in turn influence the atmospheric circulation (*Fels et al., 1980*) and therefore lead to climate changes.

Changes in the tropospheric ozone raise another environmental issue. Due to its toxic characteristics, ozone in the troposphere is regarded as a pollutant. The increase in tropospheric ozone detected at the surface (*Janach, 1989*) and in ozone sounding data (*Logan*, *1994*) has caused serious concern among scientists. Since transport of ozone from the stratosphere to the troposphere is an important source of tropospheric ozone, attention was paid to quantifying the stratospheric source, something that requires a better understanding of the processes leading to stratosphere-troposphere exchange (STE). This task is complicated by the fact that processes contributing to STE have an episodic nature.

Recently, long-term changes were also found in stratospheric water vapour, first in ground-based measurements (*Oltmans and Hofmann, 1995*), and after that in satellite data from the HALOE and SAGE II instruments (*Rosenlof et al.*, 2001). The reasons for

these changes are not completely understood, and recent studies have questioned the estimates of the trends (*Fueglistaler and Haynes*, 2005; *Randel et al.*, 2006). However, model studies motivated by the observations reveal a strong sensitivity of the temperature in the lower stratosphere to water vapour changes (*Forster and Shine, 2002*). This emphasizes needs for a better understanding of the processes controlling water vapour distribution. In particular, the relative importance for the STE of different meteorological phenomena and physical processes, especially those acting on small-scales such as convective systems, is not well-understood yet (*World Meteorological Organization (WMO), 2006*). Also, process studies in the upper troposphere/lower stratosphere (UTLS) require *in situ* water vapour measurements that were made possible only recently when lightweight ppm-grade water vapour instruments became available.

Motivated by the problems outlined above, the present work studies dynamical processes that affect the distribution of the trace gases in the stratosphere and upper troposphere. The work consists of two parts: 1) Long-term changes in the large-scale stratospheric circulation; 2) Cross-tropopause transport of ozone and water vapour in the Arctic. The objective of Part 1 is to assess long-term changes in the circulation and their contribution to ozone changes. The objective of Part 2 is to identify processes important for the two-way cross-tropopause transport of ozone and water vapour in the Arctic.

The work is based on 5 original papers. Three of them are devoted to Part 1 and the two others to Part 2. The main approach in this work is an analysis of the meteorological data from global assimilation models as well as from upper-level sounding stations. Since the largest chemical ozone losses occur in the polar regions, which through dilution also affect mid-latitudes, it is important to assess changes in the stratospheric polar vortices constituting the main feature of the wintertime stratospheric circulation. This was addressed in PAPER 3. The main forcing of the stratospheric circulation comes from the planetary waves that originate in the troposphere. They influence both the strength of the polar vortices as well as the meridional transport of substances. An analysis of the intraseasonal and interannual variability of the wave propagation into the NH stratosphere was done in PAPER 3. In PAPER 4, the changes in

the stratospheric ozone in the NH were directly related to changes in the stratospheric dynamics.

PAPERS 1 and 5 are devoted to processes at the tropopause level. In PAPER 1 the statistics of stratospheric intrusions into the Arctic troposphere was obtained from ozone- and radiosounding data. PAPER 5 focused on the tropospheric influence on the Arctic lower stratosphere. This work was based on water vapour data obtained during an Arctic field campaign at Sodankylä.

This introductory review has the following structure: The theoretical background is given in Chapter 2, sections 1-4. A brief description of the ozone and water vapour distribution in the atmosphere is given in Section 2.1. Section 2.2 gives an overview of the stratospheric meridional circulation. The meteorology of the polar vortices and the chemical processes there are reviewed in Section 2.3. Stratosphere-troposphere exchange in the extratropics is described in Section 2.4. Chapter 3 gives a short review of the results published in the papers. Chapter 4 summarises the main achievements of the work.

2. THEORY

2.1. OZONE AND WATER VAPOUR DISTRIBUTION IN THE ATMOSPHERE

The atmosphere of the Earth consists mainly of nitrogen (78.1% by volume) and oxygen (20.9% by volume), which are radiatively rather inactive. Other gases constitute only about 1% of the atmosphere's volume. However some of these trace gases, like ozone and water vapour, are radiatively very active and therefore very important for the planet's climate. The radiative balance forces the air temperature and, consequently, the atmospheric circulation of the middle atmosphere.

Neither ozone nor water vapour are uniformly distributed in the atmosphere. Figure 2.1 shows typical vertical distributions of the ozone and water vapour mixing ratios in the atmosphere. About 90% of the atmospheric ozone is contained in the stratosphere, and its mixing ratios are higher there than in the troposphere by one or two

orders of magnitude. In the stratosphere, ozone is produced through photodissociation of oxygen by ultraviolet radiation. The basic photochemical reactions naturally producing and destroying ozone in the stratosphere are known as the Chapman cycle (*Chapman*, *1930*):

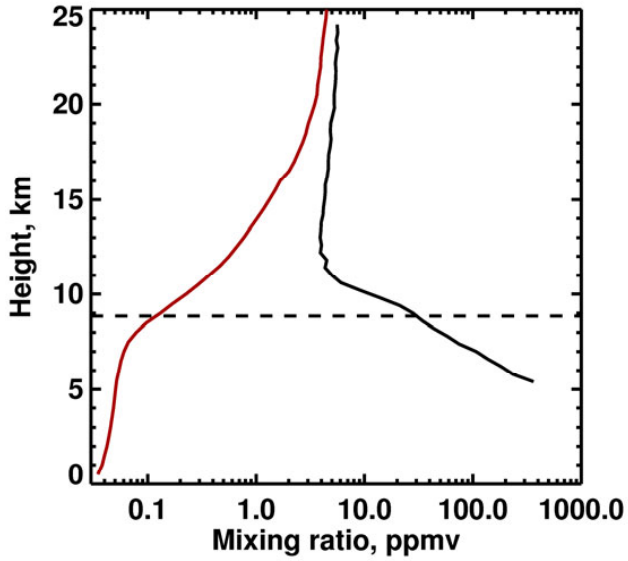

FIGURE 2.1. Mixing ratios of water vapour (black) and ozone (red) measured at Sodankylä (67.4°N, 26.6°E). The ozone climatology for February (1989-2004), and the water vapour averaged over 11 profiles obtained between 29 January and 25 February 2004 are shown. The dashed line marks the February-mean tropopause.

$$O_2 + h\nu\ (\lambda < 242\ nm) \rightarrow 2O \quad (2.1)$$

$$O + O_2 + M \rightarrow O_3 + M \quad (2.2)$$

$$O_3 + h\nu\ (\lambda < 336\ nm) \rightarrow O + O_2 \quad (2.3)$$

$$O + O_3 \rightarrow 2O_2 \quad (2.4)$$

The rate of ozone production is determined by reaction (2.1) followed almost immediately by reaction (2.2). The production rate peaks in the tropical upper stratosphere, which is a source region of ozone. Reaction (2.3) converts ozone molecule back to atomic oxygen. Because reaction (2.2) is very fast, ozone and atomic oxygen are quickly interconverted. It is convenient to regard them as a 'family' known as odd

oxygen. Reaction (2.4) is an ozone loss reaction. This reaction is however too slow and predicted ozone amount in the Tropics is too high compared to observations. Therefore, the Chapman theory was modified to include ozone losses through catalytic reactions with hydrogen (*Hampson, 1965*), nitrogen (*Crutzen, 1970*) and halogen oxides (*Stolarski and Cicerone, 1974; Molina and Rowland, 1974*). The Chapman theory also predicts ozone values in middle to high latitudes lower than observed. This disagreement can be solved if transport processes, which will be discussed in Section 2.2, are taken into account.

In the polar winter stratosphere, catalytic ozone destruction by chlorine and bromine can be strongly reinforced by heterogeneous reactions on polar stratospheric cloud (PSC) particles and result in large ozone losses. These clouds require low temperatures to form and they are observed only in the polar stratosphere. According to the current knowledge, it is the heterogeneous chemistry that is responsible for the Antarctic 'ozone hole' phenomenon. The heterogeneous chemistry will be reviewed in Section 2.3.

The chemical lifetime of ozone in the upper stratosphere (above 30km) is short. Its concentration there is determined by a balance between chemical production and destruction, so that ozone is in local photochemical equilibrium. At lower levels, the chemical lifetime of ozone becomes longer, and ozone can be transported by the atmospheric circulation. In the lower stratosphere, ozone has a chemical lifetime of weeks to months, enough for transport to play a determining role in the ozone distribution.

In the troposphere, ozone reacts quickly with nitrogen oxides, which are emitted to the atmosphere in significant amounts as a result of anthropogenic activity. In the clean, remote atmosphere, the concentration of nitrogen oxides is much lower than in the polluted atmospheres of cities and industrial centres, where chemistry can influence ozone concentration significantly. The reactions lead to either destruction or production of ozone, depending on other factors such as illumination and the presence of hydrocarbons (*Wayne*, *1991*). Also, as a strong oxidizer, ozone is effectively destroyed at the surface.

The prime source of water vapour in the atmosphere is evaporation from the surface. The water-holding capacity of air increases with temperature according to the Clausius-Clapeyron equation (*Andrews*, *2000*):

$$\frac{de_s}{dT} = \frac{Le_s}{R_v T^2}, \tag{2.5}$$

where e_s is the saturation vapour pressure, T is the air temperature, L is the latent heat of vaporization per unit mass and R_v is the specific gas constant for water vapour. Since the air temperature decreases quickly with altitude in the troposphere, nearly all atmospheric water is contained in the troposphere. In the troposphere, the vertical distribution of temperature that would be established by radiation only is unstable to convection. Vertical movements are essential in maintaining radiative-convective equilibrium. Rising air expands adiabatically and cools until the water vapour saturation pressure is achieved and condensation occurs. Hence, the water vapour distribution in the lower atmosphere is strongly influenced by phase changes.

Observations show that the stratosphere is significantly drier than the troposphere (*Brewer*, *1949*). This is related to the fact that air enters the stratosphere mainly in the tropics and passes through the cold tropical tropopause where nearly all water vapour condenses and rains down. Concentration of water vapour that passes through the tropical tropopause depends on tropopause's temperature. The effect of seasonal variation of the temperature is seen in water vapour in the tropical lower stratosphere (*Mote et al.*, *1995*). The circulation that brings air into the stratosphere and transports it therein will be discussed in Section 2.2.

Because of the significant dryness of the stratosphere very low temperatures are necessary for saturation to be achieved. Radiative cooling in winter in the polar regions leads to low temperatures; however, only in the Antarctic stratosphere are temperatures low enough for condensation to occur over large areas every winter (*Kelly et al., 1989*). In the NH, synoptic-scale ice clouds are observed only during exceptionally cold periods (*Vömel et al., 1997*, *Kivi et al., 2001*). Ice clouds are also sometimes observed at the crests of gravity waves where the adiabatic expansion of the rising air provides additional cooling (*Dörnbrack and Leutbecher*, *2001)*.

Besides evaporation from the surface, water vapour is produced in the atmosphere through certain chemical reactions. In the troposphere these contributions are negligible, but in the dry stratosphere methane oxidation contributes significantly to the water vapour budget. Water vapour production from methane oxidation is a function of altitude and latitude, but generally oxidation of each methane molecule results in approximately two water vapour molecules (*Jones et al., 1986, SPARC, 2000*). Sometimes, it is useful to consider the sum $2\times CH_4+H_2O$, referred to as 'total hydrogen' or 'potential water' (*SPARC, 2000*) since this value is nearly constant throughout the stratosphere except regions where water vapour is removed from air by dehydration, like in the polar regions, or where the water vapour concentration is modulated by tropical tropopause temperatures.

In the lower stratosphere and upper troposphere, transport appears to be important for the distribution of both water vapour and ozone. Transport contributes strongly to the seasonal variability of both quantities. In the tropopause region, where strong vertical gradients in the ozone and water vapour distributions exist, cross-tropopause transport causes strong variability on synoptic time-scales. The relevant dynamical processes will be reviewed in the following sections.

2.2 RESIDUAL CIRCULATION AND LARGE-SCALE TRANSPORT IN THE STRATOSPHERE

Many features of the global distribution of the atmospheric constituents can be understood by considering the zonally-averaged circulation, which is easier to handle mathematically than the full 3-dimensional description (*Andrews et al., 1987*). Here, the quantity A is represented as a sum:

$$A = \overline{A} + A', \tag{2.6}$$

where an overbar denotes a zonal mean that depends only on latitude, altitude, and time, and a prime denotes a departure from the zonal mean. It follows that $\partial(\overline{A})/\partial x = 0$,

$\overline{A'} = 0$, and $\overline{A} \cdot \overline{A'} = 0$, where x is the zonal coordinate. Equation (2.6) can be used to obtain a set of Eulerian-mean equations that describe flow at fixed points in 2-D latitude-altitude space (see e.g. *Andrews et al.*, *1987*). However, *Dunkerton* (*1978*) has shown that the Eulerian-mean circulation is nearly cancelled by the Stokes drift induced by eddies, and cannot therefore properly describe the transport of constituents. The small difference between the Eulerian-mean circulation and the Stokes drift is called the residual circulation, and is directly related to the mass transport. *Dunkerton* (*1978*) has also shown that the Eulerian-mean circulation and the residual circulation have opposite signs in the wintertime extratropical stratosphere and mesosphere. *Andrews and McIntyre* (*1976*) introduced a transformation that relates the residual circulation to the Eulerian-mean circulation. For β-plane geometry, the residual circulation $(\overline{v^*}, \overline{w^*})$ is defined as follows:

$$\overline{v^*} \equiv \bar{v} - \frac{R}{\rho_0 H} \frac{\partial(\rho_0 \overline{v'T'} / N^2)}{\partial z} \tag{2.7}$$

$$\overline{w^*} \equiv \bar{w} + \frac{R}{H} \frac{\partial(\overline{v'T'} / N^2)}{\partial y}, \tag{2.8}$$

where $\bar{v}$ and $\bar{w}$ are the meridional and vertical components of the Eulerian-mean circulation, R is the gas constant for dry air, H is the scale height, ρ_0 is the air density, N^2 is the Brunt–Väisälä frequency, T is the air temperature, $\overline{v'T'}$ is the eddy heat flux (HF), y is the meridional coordinate increasing northward, and z is the vertical coordinate increasing upward. The equations of mean motion for β-plane geometry can now be written as follows (*Andrews et al.*, *1987*):

$$\bar{u}_t - f_0 \overline{v^*} = \rho_0^{-1} \nabla \cdot \mathbf{F} + \bar{X} \tag{2.9}$$

$$\bar{\theta}_t + \overline{w^*} \theta_{0z} = \bar{Q} \tag{2.10}$$

$$\overline{v^*}_y + \rho_0^{-1} (\rho_0 \overline{w^*})_z = 0 \tag{2.11}$$

$$f_0 \bar{u}_z + RH^{-1} e^{-kz/H} \bar{\theta}_y = 0, \tag{2.12}$$

where t is the time coordinate, u is the mean zonal wind, f_0 is the Coriolis parameter, θ is the potential temperature ($\theta \equiv T(1000/p)^k$), X is the friction term, Q is the diabatic heating term, p is the air pressure, $k \equiv R/c_p$ where c_p is the specific heat at constant pressure, and $\mathbf{F} \equiv (-\rho_0 \overline{v'u'}, \rho_0 f_0 \overline{v'T'}/T_{0z})$ is the so-called Eliassen-Palm flux (EP flux). Zonal mean quantities are denoted by an overbar. Partial derivatives with respect to t, y, and z are denoted by suffixes. The set of equations (2.9)-(2.10) is known as the Transformed Eulerian Mean (TEM) equations.

The TEM equations can more easily be interpreted if the time derivatives in (2.9) and (2.10) may be neglected, which is a good approximation at the solstice. In the rhs of (2.9), $\rho_0^{-1}\nabla \cdot \mathbf{F}$ is the strongest contributor to forcing in the stratosphere and is mainly related to the dissipation of planetary stationary Rossby waves. The waves propagate into the stratosphere, break, and deposit momentum. This forcing causes poleward circulation through the Coriolis torque (second term in the lhs of (2.9)). This inflow of the air into the polar regions causes compression of the underlying layers, which undergo adiabatic warming. In this steady-state situation, the adiabatic warming is exactly balanced by the diabatic term $\overline{Q}$. The meridional circulation for which the adiabatic heating balances the diabatic heating associated with the observed temperature field is called the diabatic circulation (*Andrews et al.*, *1987*). From the continuity equation (2.11), sinking motion at high latitudes must be balanced by rising motion at lower latitudes.

If $\rho_0^{-1}\nabla \cdot \mathbf{F}$ is equal to zero, then the only forcing that could generate the meridional circulation would be $\overline{Q}$. In the stratosphere, the diabatic forcing is mainly due to latitudinal differences in the absorption of solar radiation by ozone. *Garcia* (*1987*) in a model study has shown that the atmosphere responds to this forcing mainly by temperature adjustment (first term in (2.10)), not by generating a meridional circulation (second term (2.10)). Wave dissipation is therefore crucial for generating the meridional circulation. This can also be illustrated by considering the angular momentum conservation law (*Haynes et al., 1991*). When the air moves from the tropics to the extratropics it loses its angular momentum, which is impossible without the

action of body forces, represented by $\rho_0^{-1}\nabla\cdot\mathbf{F}$, provided the zonal mean angular momentum distribution is steady. Only in the tropical stratosphere, where the meridional gradient of angular momentum is weak, can the differential heating contribute significantly to the driving of the meridional circulation there (*Dunkerton*, *1989*). The meridional circulation can also be driven by the seasonal transience effects as it was shown recently by *Salbi and Callaghan* (*2006*) in a model study. In this case, the annual cycle of diabatic cooling forces the transience of the angular momentum distribution.

The above paragraphs emphasize the important role of Rossby waves in generating the stratospheric meridional circulation. Consequently, the waves become important for the transport of atmospheric constituents. Following *Andrews et al.* (*1989*), the balance equation for a zonal mean chemical compound $\overline{\chi}$ in the middle atmosphere can be written as:

$$\frac{\partial\overline{\chi}}{\partial t} = -\overline{v^*}\overline{\chi}_y - \overline{w^*}\overline{\chi}_z + \rho_0^{-1}\nabla\cdot\vec{M} + \overline{S} \qquad (2.13)$$

where $\nabla\cdot\vec{M}$ is the eddy flux and $\overline{S}$ stands for chemical sources and sinks. Subscripts mark partial derivatives with respect to meridian (y) and altitude (z). The poleward mass circulation in the stratosphere is called the Brewer-Dobson (BD) circulation, and was originally deduced from observations of water vapour and ozone (*Brewer*, *1949, Dobson 1956*). Not only the residual circulation ($\overline{v^*}, \overline{w^*}$), represented by the first two terms in the rhs of (2.13), is important for transport of constituents but also the rapid quasi-isentropic transport and mixing by the breaking Rossby waves. This process is represented by the third term in the rhs of (2.13). Quasi-isentropic advection is often associated with synoptic wave events and sometimes can lead to formation of ozone minima called ‘mini-holes’ (*e.g. Hood et al., 1999; Orsolini and Limpasuvan, 2001; Koch et al., 2005*). In addition to Rossby waves, gravity waves propagating from the troposphere also contribute to forcing of the meridional circulation. It is believed that

the breaking gravity waves are the primary forcing of the meridional circulation in the mesosphere (*Andrews et al., 1989*).

Planetary stationary Rossby waves are generated at the ground by westerly flow over topography or thermal inhomogeneities throughout the year. However, only during the winter, when stratospheric winds blow eastward, can waves propagate into the stratosphere (*Charny and Drazin*, *1961*). Because of the high mountain systems and land-ocean contrasts present in the NH, the planetary Rossby waves excited at the ground are stronger there than in the Southern Hemisphere (SH). Consequently, the BD circulation is stronger in the NH. The seasonality of the wave propagation can explain the seasonal behaviour of ozone in the extratropics: the ozone content in middle and high latitudes increases in winter due to transport from the tropical stratosphere, the source region of atmospheric ozone. Moreover, the ozone buildup during the winter depends on the strength of the BD circulation. Strong correlation between the interannual variability of the BD circulation and ozone has been shown in several publications (*Fusko and Salbi, 1999; Randel et al., 2002; Hu and Tung, 2003; Nikulin and Karpechko, 2005 (PAPER 4)*). *Fusko and Salbi* (*1999*) were the first to show statistically a connection between the strength of the BD circulation and ozone. As a proxy for the BD circulation they used the HF, $\overline{v'T'}$, a quantity which is proportional to the vertical component of the EP flux. They pointed out that a negative trend in the extratropical total ozone in the NH (*e.g. Stolarski et al., 1991*) is coupled with a negative trend in the BD circulation. *Randel et al* (*2002*) have found that a weakening of the BD circulation as described by HF is restricted to the second half of winter (January-February), while there is a tendency to a strengthening of the circulation in November-December. Reasons for the difference remain unclear. *Hu and Tung* (*2003*) attempted to attribute it to the radiative forcing due to ozone trends that are confined to the late winter. *Karpetchko and Nikulin* (*2004, PAPER 2*) demonstrated a strong link between November-December HF and the circulation in January-February. They also attempted to explain January-February trends by changes in the November-December HF, but found no evidence for such a link.

The BD circulation also explains the observed dryness of the stratosphere. The rising branch of the circulation in the tropics extends through the tropopause and brings

tropospheric air containing water vapour into the stratosphere. The upwelling pushes the tropopause up making it exceptionally cold. Temperatures at the tropical tropopause decrease to below -85°C, which corresponds to a saturation mixing ratio of only 2.5-3 parts per million by volume (ppmv). On its way up to this cold point, nearly all the water vapour condenses and rains down, and the air that reaches the stratosphere is very dry. Part of the air rises into the upper stratosphere and the mesosphere where it gets additional water vapour through methane oxidation (*Jones et al., 1986*). When this air enriched with water vapour descends in the extratropics it mixes with air arriving from lower altitudes. As a result, water vapour mixing ratios in the extratropical stratosphere are higher than those in the tropics at the same altitude. Due to the methane oxidation occurring, the concentration of water vapour in the stratosphere usually increases with altitude. However, the minimum in the water vapour mixing ratio (the hygropause) in the extratropical stratosphere is located 2-3 kilometres above the tropopause. The layer between the tropopause and the hygropause gets its water vapour directly from the extratropical troposphere. The STE in the extratropics will be reviewed in Section 2.4.

2.3 THE POLAR VORTEX AND THE CHEMICAL DESTRUCTION OF OZONE

The most important feature of the stratospheric circulation at high latitudes during winter is the pole-centred cyclonical vortex. During autumn the solar irradiance in the polar stratosphere decreases quicker than that in the lower latitudes. The polar air mass gets colder and a meridional temperature gradient develops. In order to keep the thermal wind balance (2.12), westerly winds form in the stratosphere. If wave propagation from the troposphere is reduced, adiabatic warming due to the meridional circulation is weak. When temperatures inside the polar vortex drop below approximately 195K, Polar Stratospheric Clouds (PSC) consisting of frozen or partly frozen nitric acid trihydrate (NAT; Type-Ia), or of supercooled liquid ternary solution of nitric acid, sulphuric acid, and water (STS; Type-Ib) start to form. A further decrease of temperature can result in another type of PCSs consisting of ice particles (Type II) (see *Solomon, 1999*, and *WMO, 2006* for a recent review). In the case of the Antarctic stratosphere, where planetary waves are weak, PSCs of both types form every winter. In the Arctic, large-

scale PSC formation occurs only during dynamically inactive winters, when wave propagation from the troposphere is reduced (*WMO*, *2003*).

Soon after the discovery of the ozone hole in the Antarctic, it was suggested that heterogeneous reactions on the surfaces of PSC's particles play a key role in ozone hole formation (*Crutzen and Arnold, 1986; McElroy et al. 1986; Solomon et al., 1986*). Detailed discussions of the process can be found in recent reviews (*e.g. NASA 2000; WMO, 2003; WMO, 2006; Damski, 2005*).

The main source of chlorine in the stratosphere is the photolysis of man-made CFCs, which have been extensively used in industry. *Molina and Rowland* (*1974*) have shown that, since CFCs have no sinks in the troposphere, they are transported to the stratosphere where they are destroyed by solar ultraviolet radiation. For example, the CFC-12 CF_2Cl_2 is destroyed in the reaction (*Finlayson-Pitts and Pitts, 2000*):

$$CF_2Cl_2 + h\nu(\lambda<240\text{nm}) \rightarrow CF_2Cl + Cl \qquad (2.14)$$

The released *Cl* atom starts a catalytic chain reaction which leads to the destruction of O_3:

$$Cl + O_3 \rightarrow ClO + O_2 \qquad (2.15)$$

$$ClO + O \rightarrow Cl + O_2 \qquad (2.16)$$

$$\text{Net: } O_3 + O \rightarrow 2O_2$$

Because of limited amount of atomic oxygen in the lower stratosphere this process is more important in the upper stratosphere (*NASA, 2000*). According to the current understanding (*WMO*, 2006), two catalytic loss processes, first proposed by *Molina and Molina* (*1987*) and *McElroy et al.* (*1986*), are mainly responsible for the polar ozone depletion:

Cycle 1:

$$ClO + ClO + M \rightarrow Cl_2O_2 + M \qquad (2.17)$$

$$Cl_2O_2 + h\nu \rightarrow 2Cl + O_2 \qquad (2.18)$$

$$2\times(Cl + O_3 \rightarrow ClO + O_2) \quad (2.19)$$

Net: $2O_3 \rightarrow 3O_2$

Cycle 2:

$$BrO + ClO \rightarrow Cl + Br + O_2 \quad (2.20)$$

$$Br + O_3 \rightarrow BrO + O_2 \quad (2.21)$$

$$Cl + O_3 \rightarrow ClO + O_2 \quad (2.22)$$

Net: $2\ O_3 \rightarrow 3\ O_2$

The reactive chlorine compounds *Cl* and *ClO* are typically removed from the atmosphere through the reactions with methane or with nitrogen dioxide:

$$Cl + CH_4 \rightarrow HCl + CH_3 \quad (2.23)$$

$$ClO + NO_2 \rightarrow ClONO_2 \quad (2.24)$$

The release of chlorine from *HCl* and $ClONO_2$ through gas-phase reactions is very slow, so these compounds act as reservoirs for chlorine. Under normal stratospheric conditions, chlorine released from CFCs is mainly contained in these two reservoir compounds and the catalytic reactions described above cannot result in significant ozone losses (*WMO*, 2006). In order to initiate the catalytic ozone loss processes chlorine should be converted into reactive form. *Solomon et al* (*1986*) suggested that chlorine can be quickly released from the reservoirs through reactions on the surfaces of PSC particles:

$$HCl\ (s) + ClONO_2\ (g) \rightarrow Cl_2\ (g) + HNO_3\ (s) \quad (2.25)$$

In the presence of sunlight, Cl_2 is quickly dissociated, producing two chlorine atoms which initiate the ozone-destroying cycle. The presence of sunlight is therefore crucial for ozone destruction to take place. As nitric acid HNO_3 remains in the solid state it sediments from the stratosphere with PSC particles. This results in a removal of nitrogen from the gas phase, a process known as denitrification. Denitrification prevents

formation of the reservoir $ClONO_2$ and increases ozone loss. In addition to reaction (2.25), there are other heterogeneous reactions which convert chlorine into reactive forms (*see e.g. NASA, 2000*).

Ozone is supplied into the polar vortex by descending branch of the BD circulation. This descend is relatively unmixed as strong zonal winds at the edge of the vortex prevent mixing of the polar air mass with air from outside the vortex (*Chen, 1994*). Weaker circulation results in less ozone supply and also leads to colder temperatures that favour formation of PSCs. Heterogeneous reactions on PSC particles convert chlorine into reactive form, which gives start to catalytic ozone-destroying reactions once sunlight is present. Weak mixing across the vortex edge keeps the concentration of ozone-destroying compounds high enough for effective ozone destruction to take place and also limits the quasi-isentropic ozone transport from midlatitudes. In the Antarctic, this results in a nearly complete ozone removal by the time of vortex breakup at altitudes ~14-22 km where PSCs exist (*WMO, 2003*). In the Arctic, significant ozone losses have been observed only during exceptionally cold winters. The first evidence of ozone depletion in the Arctic was obtained from the AASE campaign in the winter of 1988/1989 (*Hofmann et al., 1989, Kyrö et al., 1992*). Later, the ozone depletions in the Arctic springtime have been accurately quantified by ozone sonde techniques (*von der Gathen et al., 1995; Rex et al., 1997*).

The above paragraphs show that ozone losses strongly depend on the abundance of PSCs. Hence, they depend on the dynamical factors that regulate temperatures inside the polar vortex, as well as the strength of the vortex (measure of impermeability), the vortex area and its longevity. The interannual variability of the vortices is linked to several processes such as the solar cycle (*Labitzke, 1987*), the quasi-biannual oscillation, QBO (*Holton and Tan, 1980; Garcia and Solomon, 1987*) and the El-Nino Southern Oscillation, ENSO (*Hamilton, 1993; Baldwin and O'Sullivan, 1995*). However, the global meridional circulation generated by Rossby waves is believed to be the most important factor, especially in the NH (*Waugh et al., 1999*). The meridional circulation keeps the temperature inside the vortex higher than the radiative equilibrium temperature (the temperature determined by a balance between the absorption and emission of radiation). It has been shown that strong correlations exist between the

strength of the meridional circulation represented by the vertical component of the EP flux and the zonal mean temperatures north of ~60°N (*Newman et al., 2001*), the longevity of the Arctic vortices (*Waugh et al., 1999*), the area with temperatures lower than the threshold temperature for PSC formation (*Weber et al., 2003*) and the vortex area and strength (*Karpetchko et al (2005, PAPER 3*)).

2.4 STRATOSPHERE-TROPOSPHERE EXCHANGE (STE)

Downward transport through the tropopause is an important source of tropospheric ozone. On the opposite, upward transport of water vapour through the tropopause is the dominant source of lower stratospheric water vapour. Both ozone and water vapour are important for the radiative state of the upper troposphere-lower stratosphere (UTLS) region (*Forster and Tourpali, 2001*). Calculations by *Forster and Shine* (*2002*) indicate that the water vapour trends reported by *Oltmans et al.* (*2000*) may have resulted in a cooling of the lower stratosphere of 0.8 K over two decades. Ozone and water vapour are also crucial for the chemical balance of the UTLS region. *Esler et al.* (*2001*) have shown that the mixing of stratospheric ozone and tropospheric water vapour results in an increased production of the hydroxyl radical, which is one of the main oxidants in the atmosphere.

Figure 2.1 shows strong gradients in both the ozone and the water vapour distribution at the tropopause, suggesting that vertical transport across the tropopause is weak. The restriction of cross-tropopause transport is related to a strong vertical gradient of a quantity called potential vorticity (PV). The expression for PV can be written as (*Hoskins et al.*, *1985*):

$$\mathrm{PV} = -g(\zeta + f)\partial\theta/\partial p, \tag{2.26}$$

where PV is the potential vorticity, g is the acceleration due to gravity and ζ is the vertical component of the relative vorticity. The unit of 10^{-6} K m^2 kg^{-1} s^{-1} is called a potential vorticity unit (PVU). Tropospheric values are generally below 1.5 PVU. In the stratosphere, PV increases steeply with height. Values between 1.5 and 4 are usually

used to define the tropopause. The tropopause defined in this way is called the dynamical tropopause. It can be shown that PV is a conserved quantity in adiabatic, frictionless flow (*Hoskins et al.*, *1985*). Since cross-tropopause transport is associated with changes in PV, STE is constrained by the rate at which diabatic and frictional processes occur.

Figure 2.2 shows schematically the major transport pathways in the UTLS region (*from Stohl et al., 2003*). The BD circulation includes the rising motion in the tropics and the sinking motion in the extratropics. As was pointed out in Section 2.2, this circulation is a diabatic one. The tropical tropopause is located on the $\theta \approx 380$K surface and declines towards the poles to $\theta \approx 290$-300K. Tropospheric air can reach the stratosphere higher than $\theta \approx 380$K only in a slow diabatic motion. At altitudes lower than $\theta \approx 380$K, STE can occur along isentropic surfaces. The part of the stratosphere below $\theta \approx 380$K is called the lowermost stratosphere.

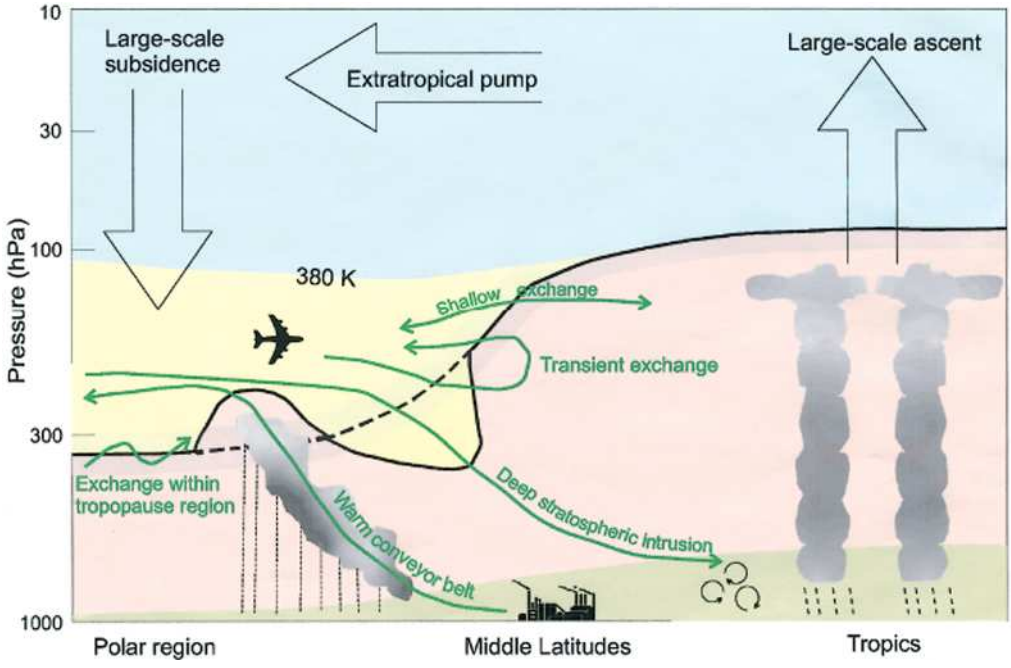

FIGURE 2.2. Schematic picture of stratosphere-troposphere exchange. Shown are the mean position of the tropopause and the main pathways of the exchange. (from *Stohl et al., 2003*. Reproduced with permission of the American Meteorological Society.)

Several processes leading to STE in the extratropics have been so far identified. Intensive quasi-horizontal STE, also referred to as 'quasi-isentropic', occurs near the jet stream (*Hoerling et al., 1993*). There, STE may be associated with strong turbulence, which provides conditions for the non-conservation of PV (*Shapiro, 1980*). STE near the jet stream is facilitated by the strong three-dimensional deformation of the tropopause associated with tropopause folds (*Danielsen et al., 1970*) and filamentation (*Appenzeller and Davies, 1992; Appenzeller et al., 1996a*). STE also occurs as a result of tropopause erosion by convective clouds. Convection is accompanied by the release of latent heat which leads to strong diabatic heating. Tropopause erosion has been observed inside a mesoscale convective complex (*Poulida et al., 1996; Rood et al., 1997*) and inside cut-off cyclones (*Price and Vaughan, 1993; Gouget et al., 2000*).

Though the global circulation in the extratropics is directed downwards, measurements of trace gases in the UTLS have shown that STE in the extratropics takes place in both directions. As a result of the two-way transport and subsequent mixing, a layer consisting of a mixture of stratospheric and tropospheric air forms around the tropopause (*Ray et al., 1999, Fisher et al., 2000, Pan et al., 2004*). *Karpechko et al* (*2006, PAPER 5*) used balloon-borne measurements of water vapour and ozone to describe the characteristics of this layer in the wintertime Arctic lowermost stratosphere.

Holton et al. (*1995*) argued that the global mass flux through the tropopause does not depend on the details of the exchange at the tropopause level. *Appenzeller et al.,* (*1996b*) suggested a method in which the budget of the whole lowermost stratosphere is considered. The STE is calculated as the difference between the mass change of the lowermost stratosphere and the mass flux through the 380K isentropic surface. *Schoeberl* (*2004*) advanced this method by separating the isentropic and diabatic parts of the STE. These studies revealed a pronounced annual cycle in the extratropical STE with downward flux being largest in late winter/spring and weaker in late summer/autumn. However two-way flux estimates obtained from different data sets show significant disagreement (*Schoeberl, 2004*). Since the net flux from the stratosphere to the troposphere is the residual of upward and downward fluxes, details of the exchange at the tropopause level are becoming important. Therefore, methods to be applied at the tropopause level are required.

One such method includes the statistical analysis of intrusions from tracer measurements (*van Haver et al., 1996*). *Karpetchko et al* (*2003, PAPER 1*) used this method to obtain statistics of ozone intrusions into the Arctic troposphere. Another approach is the direct calculation of mass fluxes from meteorological data. *Wei* (*1987*) derived a formula for flux calculation in a generalized vertical coordinate. However, when pressure or isentropic temperature are used as a vertical coordinate, the results are highly inaccurate, since flux is calculated as the residual of large terms that nearly compensate each other (*Wirth and Egger, 1999*). This problem disappears when PV is used as a vertical coordinate. The results obtained with PV-coordinates are in good agreement with other methods, such as the Lagrangian approach, in which the mass flux is obtained from trajectory calculations (*Wernli and Borqui, 2001*). In the case of PV-coordinates, the Wei formula takes the form:

$$F = -\frac{1}{g}\frac{\partial p}{\partial (PV)}\frac{D(PV)}{Dt} \tag{2.27}$$

Sigmond et al. (*2000*) suggested calculating the material derivative of PV along trajectories. Here, trajectories initiated at the tropopause are run forward and backward in time and $D(PV)/Dt$ is calculated as the difference between the PV values at the ends of the trajectories divided by the time interval. *Karpechko et al.* (*2006, PAPER 5*) used (2.27) in a case study to analyse the STE associated with a mid-latitude cut-off anticyclone in winter.

3. RESULTS

3.1 STRATOSPHERIC DYNAMICS AND LONG-TERM CHANGES IN THE STRATOSPHERIC CIRCULATION

PAPER 2 was stimulated by a paper by *Randel et al.* (*2002*), who found opposite HF trends between November-December (ND) (positive trend) and January-February (JF) (negative trend) in the NH. What causes the opposite trends remains unclear. Some models predict reduced wave propagation to the stratosphere as a result of the increase in greenhouse gases (*Shindell et al., 1999*) while others report enhanced wave propagation due to the increase in greenhouse gases (*Butchart et al.*, *2001*).

The investigation into wave behaviour performed in PAPER 2 revealed that the HF in ND and JF are strongly anticorrelated. A strong wave flux into the stratosphere in early winter is usually followed by a weak flux in midwinter and vice versa. Figure 3.1 shows the correlation between the ND total HF at 20hPa averaged over 45-75°N and the JF total HF for the period 1979-2006. This plot is an update of the original result obtained from a shorter time-series (1979-2002). Comparison of Fig. 3.1 with Fig. 1 of PAPER 2 shows that the addition of 4 years does not change the picture significantly. The robustness of the result was also confirmed in a longer time-series (1958-2002). Three centres of statistically significant correlation can be identified: 1) negative correlations in the middle and high latitude stratosphere and 2) in the troposphere, and 3) a positive correlation in the subtropical lower stratosphere. Spectral analysis showed that the first centre is associated with the JD HF due to planetary waves 1-3; the third centre is associated with JD HF due to short waves 4-7; and the second one is associated with the JD total HF.

The response in the stratosphere is believed to be a manifestation of stratospheric vacillation cycles like those found in a quasigeostrophic β-plane channel model by *Holton and Mass* (*1977*). Here, strong wave flux leads to a deceleration of the westerly winds, the establishment of easterlies, and, subsequently, a reduction of wave flux, with the period of vacillation being determined by radiative relaxation times. The

anticorrelation is also consistent with the suppression of wave propagation due to the observed decrease in wind shear at the tropopause level, a relation that follows from linear wave theory (*Hu and Tung*, *2002*). The two other centres are not so easy to interpret. Speculation presented in the paper remains so far without proper testing, which ought to involve model runs.

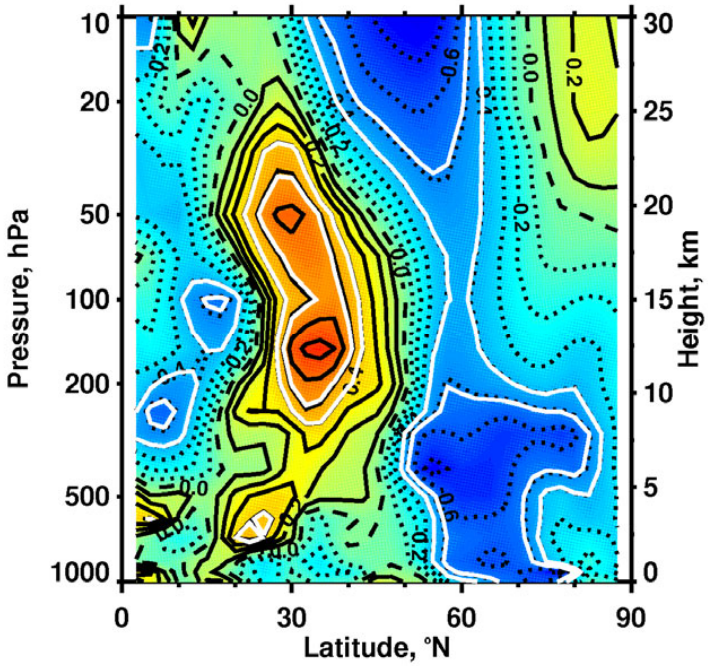

FIGURE 3.1. Correlation between the November-December heat flux at 20hPa averaged over 45-75°N and the January-February heat flux (1979-2006) in the Northern Hemisphere. Positive contour values are solid, negative values are dotted and zero contours are dashed. The first and second white lines are the 95% and 99% confidence levels (Courtesy of G. Nikulin).

However, the mechanisms responsible for the anticorrelation between ND and JF HF in the stratosphere are hardly responsible for the opposite trend behaviour in these seasons reported by *Randel et al* (*2002*). A latitude-altitude cross-section of the trends is shown in Figure 3.2. While the JF stratospheric response to ND HF is located mainly south of 60°N (see Fig. 3.1), the JF negative trends are shifted northward of 60°N, with statistically significant trends located mainly in the lower stratosphere, where the correlation is barely or not at all significant. In the upper troposphere, the area of

significant trends only partly overlaps the area of negative correlation. It is known from linear wave theory that wave propagation depends on the zonal wind speed and its vertical shear. Thorough inspection reveals no significant trends in either of these parameters in the period 1979-2002. This makes a physical interpretation of the HF trends difficult. Also, trends in the NH stratosphere are strongly sensitive to the period chosen, due to the large interannual variability.

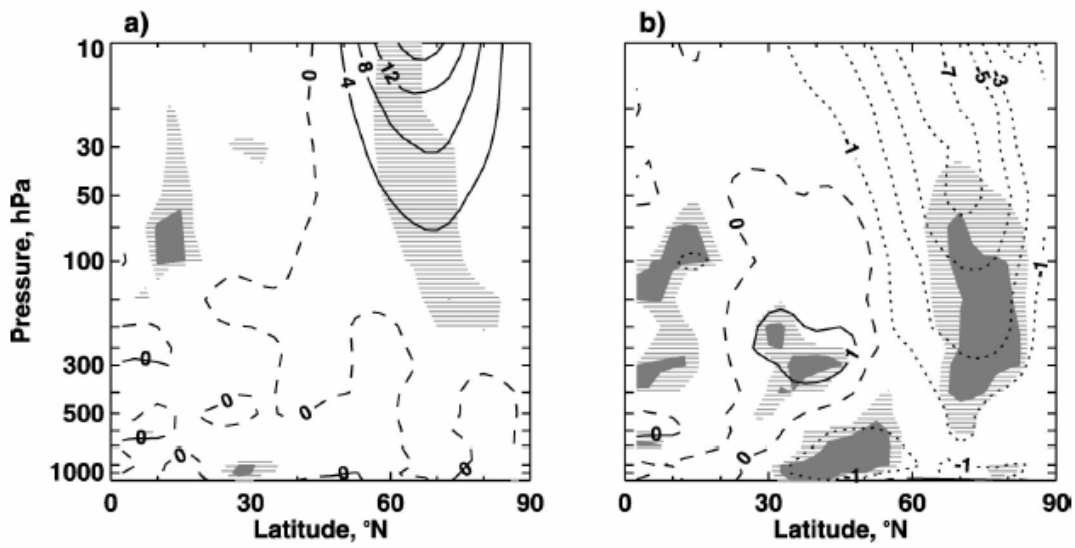

FIGURE 3.2. Decadal trends in heat flux averaged for (a) ND and (b) JF for 1979–2002. Units are in K m/s per decade. Positive contour values are solid, negative values are dotted, and zero contours are dashed. The 95% (lines) and 99% (solid) confidence levels are shaded. (From *Karpetchko and Nikulin (2004)*. Reproduced with permission of the American Meteorological Society.)

The long-term changes in the stratosphere were further assessed in PAPER 3 focused on the polar vortices in both hemispheres. Data from ECMWF ERA-40 reanalyses covering the period from 1957 to 2002 were initially chosen for the study. First, a quick data quality control was performed, since this new data set had not been previously used for similar studies. This was done by comparing the ERA-40 time-series with those from NCEP/NCAR. Although the NCEP/NCAR reanalyses have their weaknesses (*Kistler et al.*, *2001*), such a comparison makes sense, since the NCEP/NCAR data have been widely used in many previous vortex studies. Analysis revealed vertical

oscillations in the ERA-40 temperature, mainly above 10hPa but also, to a smaller extent, in the lower stratosphere. The largest differences between the two datasets were found in the polar stratosphere of the SH. Some of the differences were attributable to problems with the assimilation of satellite data in ERA-40. Similar results were concurrently reported by *Renwick* (*2004*) and *Randel et al.* (*2004*). As a result of the quality tests, only data after 1979 were eventually used in studies of the Antarctic vortex.

Several vortex parameters, such as vortex area, strength, mean temperature, longevity and PSC area, were calculated on seven isentropic levels (395K, 430K, 475K, 530K, 600K, 700K, 850K). The analysis included a study of the parameter interrelations and long-term variability. In the NH, all vortex parameters were found to be strongly correlated with each other. For example, a larger vortex is usually stronger and colder than a smaller one. All the parameters depend strongly on the HF into the stratosphere integrated over the preceding period of 1-2 months. The HF determines the time of vortex break-up, a result also obtained earlier by *Waugh et al* (*1999*). The significant correlation found between the HF and the PSC area has an implication for statistical ozone models. Since the PSC area is regarded as a proxy for the chemical ozone depletion inside the vortex (*Rex et al.*, *2004*), it is important to realize that about 50% of its interannual variability in the NH is explained by the variability of the planetary wave flux. Simultaneous use of the PSC area and HF in statistical models should therefore be made with caution. A similar relationship between the PSC area and the HF was also obtained by *Weber et al.* (*2003*) on a shorter timescale.

A statistically significant correlation between the PSC area and the HF is also found in the SH polar stratosphere in winter. However, the correlation disappears by October. This is related to the different long-term behaviour of the parameters. While the HF shows virtually no changes over the period 1979-2001, a significant increase is observed in the springtime PSC area. When the increase is filtered out, the statistically significant correlation is restored. This suggests that the increase in the PSC area is not caused by changes in the circulation. Presumably, the increase is related to the cooling of the Antarctic stratosphere due to ozone depletion (*Randel and Wu, 1999*).

TABLE 3.1. Trends and two-sigma uncertainties in vortex parameters. Trends significant at the 95% level are marked in blue

	Isentropic level, (K)	NH			SH
		(1958-2002)	(1979-2002)	(1979-1997)	(1979-2001)
Break-up date, (days/decade)	475	0.8±3.8	5.0±11.0	15.1±12.7	9.8±5.7
	600	1.1±4.1	3.7±12.2	11.2±13.9	9.1±5.7
	850	-0.9±3.6	7.5±9.1	10.7±12.4	5.1±5.8
March(NH)/October(SH) vortex strength, ((m/s)× (PVU/deg)/year)	475	0.3±0.7	1.0±2.0	3.7±2.2	4.1±2.0
	600	1.2±1.6	2.8±4.7	8.2±5.7	5.7±3.3
	850	-0.6±0.9	0.9±2.0	2.9±2.9	2.1±1.7
March(NH)/October(SH) vortex area (10^6 km^2/decade)	395	2.2±3.0	1.7±8.7	14.4±10.0	0.1±2.7
	475	0.7±2.1	0.8±5.8	7.5±7.0	-1.8±2.7
	530	0.5±2.1	0.5±5.6	6.8±6.7	-1.6±2.8
March(NH)/October(SH) PSC area (10^6 km^2/ decade)	395	0.16±0.15	0.4±0.5	0.9±0.7	3.8±1.8
	475	0.15±0.15	0.3±0.5	0.8±0.7	3.1±1.1
	530	0.06±0.07	0.1±0.3	0.4±0.4	0.2±0.5

Table 3.1 summarises the results of the trend calculations for the vortex parameters. The results are shown for springtime. This is the time when the polar stratosphere is exposed to sunlight which can initiate ozone-destroying catalytic reactions explained in Section 2.3. It is seen that none of the Arctic vortex parameters exhibit a significant trend in the period 1979-2002. The trends are significant only if the period 1979-1997 is considered. The second half of the latter period is marked by several exceptionally cold winters in the Arctic stratosphere that were accompanied by severe ozone losses. If the whole ERA40 period is considered, statistically significant trends are observed only for the PSC area. Though the trends found here are just barely significant at the 95% confidence level, an increase in the PSC area has also been reported by other authors (*Knudsen et al., 2004, Rex et al. 2004*). *Rex et al.* (*2004*) noticed that the increase

becomes more pronounced only if the coldest winters from 4-10 year intervals are considered.

In the Antarctic, the springtime vortex in the lower stratosphere shows statistically significant trends in temperature, longevity and strength in the period 1979-2001. As was mentioned above, these changes cannot be related to changes in the circulation. Another interesting result of the paper is that no long-term changes are found in the area of the Antarctic vortex at any of the levels studied. This is consistent with the findings of *Bodeker et al* (*2002*) at the 550K isentropic level.

Transport of ozone to the extratropics by the BD circulation in the NH winter was studied in PAPER 4. The paper was focused on ozone tendencies ($\Delta O_3/\Delta t$) rather than on ozone values, since it is the ozone tendency that is directly related to the meridional circulation, as can be deduced from Eq. (2.13). During summer, ozone tendencies in the extratropics are negative due to photochemical destruction (*Fioletov and Shepherd*, *2003*). The results of PAPER 4 suggest that positive ozone tendencies start in October over the Pacific Ocean, spread zonally in November, and completely cover mid-latitudes in December. To the writer's knowledge, the strong zonal asymmetry during the early stage of the ozone buildup had not been mentioned earlier in the literature. The spatial structure of the ozone tendencies from November to January resembles that of stationary planetary waves, but in February and March the differences become significant. This suggests that dynamics dominate the ozone changes during the former period, while other factors, presumably ozone-related chemistry, become equally important during the latter.

The HF is traditionally used as a proxy of the wave forcing of the meridional circulation (*Fusko and Salbi, 1999; Randel et al. 2002*) because this parameter is easy to calculate. It is common practice to use the HF at 100hPa averaged over a latitude range (e.g. 45°-75°N). However, the maximum in the correlation between HF and ozone tendencies is usually located above 100hPa (see Fig. 3, PAPER 4). The HF at 100hPa is thus not always an optimal proxy for the ozone tendency. The correlation pattern between $\Delta O_3/\Delta t$ and HF changes from month to month. For example in December, the maximum correlation coefficient is rather modest (r_{max}=0.45). By contrast, the correlation pattern between $\Delta O_3/\Delta t$ and the vertical component of the residual

circulation $\overline{w^*}$ changes only little from month to month. On average, the correlation between $\Delta O_3/\Delta t$ and $\overline{w^*}$ is close to -0.7 for all months from November to March.

According to the results of PAPER 4, the interannual variability of the BD circulation explains about 50% of the interannual variability of extratropical ozone for the period 1980-2002. This estimate is lower than that obtained by *Salby and Callaghan*, (*2002*) and *Weber et al*. (*2003*), but higher than that obtained by *Randel et al*. (*2002*).

3.2. INFLUENCE OF THE STE ON THE DISTRIBUTION OF TRACE GASES AT THE ARCTIC TROPOPAUSE

The influence of transport on the gas distribution in the tropopause region in the Arctic was studied in PAPERS 1 and 5. PAPER 1 gives statistics for stratospheric intrusions into the troposphere based on ~10 years of ozone and radiosoundings at two Arctic stations: Sodankylä (67.4°N, 26.7° E) and Ny-Ålesund (78.9° N, 12.0° E). To detect stratospheric intrusion into the troposphere, the algorithm based on that described in *van Haver et al.* (1996) was developed. In this method, intrusion is identified as a dry, stable and ozone-rich layer well-detached from the stratosphere. The requirement of dryness excludes detection of layers with photochemically-produced ozone from the lower troposphere.

The method was originally developed for the detection of a tropopause fold, which is a lowering of the tropopause on the cyclonic side of the jet stream resulting from the ageostrophic circulation (e.g. *Shapiro*, *1981*). The tropopause fold is a reversible process. In the entrance region of the jet stream the ageostrophic circulation is thermally direct and air sinks on the cyclonic side. In the exit region the ageostrophic circulation changes to become thermally indirect. As a result, a large part of the descended air returns back into the stratosphere.

Practice has shown that stratospheric layers are sometimes observed away from the jet stream, where vertical movements are weak. Such layers are most probably remnants of earlier intrusions and have experienced prolonged mixing with tropospheric air (*Bithell et al., 2000, Vaughan and Worthington, 2000*). They are therefore of more

interest for the STE. To obtain statistics of these intrusions (thereafter referred to as 'aged') additional criteria were added to the original method to automatically exclude layers near the jet stream. If the original strict criteria are used, aged layers are detected in 2% and 3% of all soundings at Sodankylä and Ny-Ålesund respectively. This is approximately twice as infrequent as the frequency of occurrence of a tropopause fold (6% at Sodankylä and 5% at Ny-Ålesund). Since the original criteria were developed for the detection of fresh intrusions, they may rule out aged layers exposed for a longer time to non-conservative processes and therefore less pronounced. According to case studies, the characteristic age of the layers detected is 2-4 days. *Foster and Wirth* (*2000*) have shown that a static stability anomaly, which is part of a PV anomaly, decays to some 70% of its initial strength after 3 days. Relaxing the original criteria by 30%, occurrence frequencies of 6% for Sodankylä and 7% for Ny-Ålesund were obtained. These values are close to the frequencies of tropopause folds, suggesting that there might be a link between the processes. Trajectory calculations performed for one event showed that the stratospheric air descended to tropospheric altitudes inside a tropopause fold and was subsequently transported away from the jet stream during the poleward breaking of a synoptic Rossby wave.

A larger part of aged layers is observed in this study north of the jet stream and is associated with streamers developed during poleward breaking of synoptic Rossby waves. This contrasts to the situation in mid-latitudes, where intrusions are typically associated with the equatorward breaking of synoptic Rossby waves, as observed from PV maps and satellite water vapour images (*Appenzeller et al., 1996a*).

PAPER 5 is devoted to the analysis of the water vapour distribution in the Arctic wintertime lowermost stratosphere. The work is based on 11 profiles obtained between 30 January and 25 February 2004. The concentration of water vapour exhibits a dramatic change across the tropopause. At the tropopause, water vapour mixing ratios vary from 10 to 40 ppmv, but decrease rapidly with height to typically stratospheric values of about 4-5 ppmv (see Figure 1 of PAPER 5). The upper level at which flight-to-flight variability was observed was located 2.5 km (or 30K on a potential temperature scale) above the local tropopause. These estimates are reasonably close to results obtained earlier from aircraft measurements (*Pfister et al., 2003, Hoor et al. 2004*). The

cross-correlation of water vapour with ozone shows that the layer characterised by flight-to-flight variability contains air with a chemical composition intermediate between tropospheric and stratospheric. This layer is called the mixing layer (*Hoor et al. 2004*) or transition layer (*Pan et al., 2004*). The concentration of water vapour continues to decreases with height above the mixing layer, and the hygropause, the level of the water vapour minimum, was located at 365K. This is approximately 3 km higher than the altitude of the tropopause during the observations. Since there are no sources of water vapour in the stratosphere at these altitudes, the hygropause marks the upper level at which transport from the extratropical troposphere takes place. In fact, there are observational evidences for transport of tropospheric air up to 360–380K in winter (*Vaughan and Timmis, 1998*).

It is important to identify the transport processes that regulate the water vapour distribution in the UTLS. Layers of enhanced water vapour in the stratosphere were of particular interest in the study, since a laminated structure in the vertical profiles of trace gases that have a strong gradient across the tropopause is usually indicative of recent transport. To identify the origin of this moist air, backward trajectories were run clustered around the station. Analyses of the trajectories and global meteorological fields showed that the observed layers were related to filamentation around upper-level cut-off anticyclones. However, filamentation itself is a purely dynamical process resulting from non-linear effects in 2-D flow (*Polvani and Plumb*, *1992*). It is not associated with changes in PV and therefore can not account for transport across the dynamical tropopause. As was discussed in Section 2.4, STE can be associated either with diabatic processes or with turbulence. Mid-latitude cut-off anticyclones are not usually regarded as important agents of STE (*Hoskins et al., 1985*). Convection in anticyclones is suppressed and erosion of the tropopause by convection is unlikely. Radiative processes are slow and cannot efficiently influence the anticyclone during its lifetime (*Hoskins et al.*, *1985*). On the other hand, clear-air turbulence (CAT) can form in the region of strong winds surrounding the anticyclone. Analysis of the turbulence index (which is a diagnostic for CAT (*Ellrod and Knapp 1992*) along the backward trajectories performed in PAPER 5 suggested a link between CAT and upward transport across the tropopause. Further, calculation of the cross-tropopause fluxes with Eq.

(2.23) showed that the surroundings of cut-off anticyclone were indeed a region of intensive two-way STE. In such a region, an air mass that moves from the troposphere to the stratosphere can easily return to the troposphere. However, if the air mass is transported away by a filament, as was observed in our study, it more probably contributes to irreversible STE.

Though, statistically, a link between the turbulence index and the upward cross-tropopause fluxes turned out to be rather weak during the period studied, the results of PAPER 5 suggest that development of a mid-latitude cut-off anticyclone, the turbulence at its flanks and subsequent filamentation may be an important mechanism for the moistening of the lowermost extratropical stratosphere.

4. CONCLUSIONS

This dissertation studies certain dynamical processes important for the distribution of ozone and water vapour. Two problems are considered: 1) Long-term changes in the large-scale stratospheric circulation and 2) Cross-tropopause transport of ozone and water vapour in the Arctic. Though the problems are considered separately, both of them stem from a more general problem: the separation of natural and anthropogenic changes in radiatively-important atmospheric gases.

Long-term changes in extratropical stratospheric ozone are driven by dynamics and chemistry, the latter being largely dependent on the former. The interannual seesaw between the early and late winter HF, the driving force of ozone transport, is revealed in the extratropical stratosphere and interpreted as an internal oscillation in the wave-mean flow system (PAPER 2). The oscillations are not however linked to opposite HF trends between early and late winter months. These trends, found in earlier studies and confirmed in the present study, are difficult to interpret, since related quantities such as mean zonal wind and vertical wind shear show no systematic changes for the same period. Also, no statistically significant long-term trends are found in the parameters of the Arctic polar vortex (PAPER 3). Anomalously cold and persistent polar vortices observed in the Arctic during 90s have become rarer during recent years. These changes are mirrored in the behaviour of the extratropical ozone north of about 30°N. The negative ozone trends observed in the NH during the previous two decades have reversed in the mid-1990s (*Andersen et al., 2006, Weatherhead and Andersen, 2006 and references therein*). Some recent studies suggest that the turnaround in ozone trends in NH mid-latitudes is linked to changes in dynamics (*Hadjinicolaou et al., 2005*), while others attribute at least part of the change to the positive effect of the Montreal protocol and its amendments (*Yang et al., 2006*). According to estimates presented here, the dynamical variability accounts for about 50% of the ozone variability in NH mid-latitudes (PAPER 4). At the same time, significant long-term changes are found in the Antarctic polar vortices, which get colder and stronger.

The transport of ozone and water vapour in the Arctic UTLS was studied using data from sounding stations. The results of the dissertation demonstrate the importance

of poleward Rossby wave breaking for STE in the Arctic. Poleward Rossby wave-breaking appears on synoptic charts as tropospheric streamers or cut-off anticyclones. The development of these systems is often accompanied by significant distortion of the background chemical composition below and above the tropopause. This is supported by the statistics of ozone intrusions into the troposphere (PAPER 1) and the analysis of water vapour distribution above the tropopause (PAPER 5). Also, calculations of cross-tropopause fluxes suggest that intensive two-way STE takes place around cut-off anticyclones. Part of this STE is caused by clear-air turbulence, as shown in PAPER 5.

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