# Overview and Results of THESEO 1998-1999

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**SUMMARY**

The Third European Stratospheric Experiment on Ozone (THESEO) took place during 1998 and 1999. The main aim was to improve our understanding of the causes of ozone depletion over Europe and other mid-latitude regions, where the ozone layer has been progressively depleted for the last 20-30 years. In order to achieve this four areas of scientific enquiry were defined:

- the mid-latitude lower stratosphere
- the links between the mid-latitude lower stratosphere and the surrounding atmosphere
- the Arctic vortex
- the tropics and sub-tropics

In this summary, we present the main findings to date which relate to these four areas.

*Mid-latitude lower stratosphere*

In addition to a strong seasonal cycle and a long term decline, the observational record of ozone over mid-latitudes shows pronounced interannual variability. This feature has received a lot of attention recently as it is necessary to describe the variability correctly in order to calculate the trends accurately. Studies of long term measurements have shown a strong link between total ozone over Europe and tropospheric circulation features such as the Arctic Oscillation. These imply that the

halogen-induced part of the ozone trend over Europe is less than would be found without taking these factors into account. The existence of such a link also explains to some degree the relatively high ozone values seen at mid and high latitudes in 1998 and 1999. The possible connection of the long term changes in the Arctic Oscillation with climate raises the possibility that this is a mechanism through which climate change will affect stratospheric ozone. Other long term records show that stratospheric aerosol amount in 1999 was as low as in the late 1970s and that the stratospheric halogen loading is peaking in the low stratosphere in 1999/2000.

The relative importance of the causes of middle latitude ozone decline remains controversial. It has been argued by some that much of the loss occurs *in situ* while others have suggested that polar processes have a significant influence on the decline of ozone in mid-latitudes. Preliminary analyses of ozone measurements in THESEO provide the first direct observational evidence that ozone loss occurs over mid-latitudes and, further, that the ozone loss is faster at lower temperatures. Together these analyses indicate the importance of the cold regions which often lie across the vortex edge. Modelling studies of where ozone depletion occurs and where ozone depleted air ends up suggest that in both THESEO winters, the disturbed stratosphere resulted in ozone loss being spread over large areas. This contrasted with some earlier years when a strong vortex encompassed the cold region and large ozone losses were confined to the vortex.

Bromine compounds contribute to the chemical depletion of ozone at mid-latitudes. Studies in THESEO have shown that stratospheric bromine loading is about 20 ppt, up to 25% higher than previously thought. Further, there is good agreement between observed and modelled BrO concentrations. As a result of these advances, our overall understanding is now such that we have confidence that current models can be used to assess the role of bromine in mid-latitude ozone loss.

*Linkages*

The two THESEO winters were relatively disturbed and have provided a good opportunity to investigate transport and mixing. Significant erosion of the Arctic vortex occurred during the major midwinter warming in December 1998 and in February 1999. In the latter case two-way mixing of air between the vortex and mid-latitudes was observed and has been reproduced in models. Air masses with different chemical compositions were mixed, resulting in net ozone loss. During spring 1999, the air over Europe was a complicated mixture of air (as seen in $H_2O$ and long-lived tracers) from the vortex, the sub-tropics and mid-latitudes. Case studies indicate that mixing rates of vortex filaments into mid-latitudes are a few weeks. For the first time, intensive observations of ozone around the sub-tropical jet were made in both hemispheres during winter. Large scale filamentary exchange of air between the troposphere and stratosphere was clearly observed in the ozone vertical cross-sections. A regional model with a 50 km grid and a high resolution advection model are able to reproduce the observations and are being used for air mass transport estimates.

Investigations of the composition of air in the lowermost stratosphere and upper troposphere were made in July 1998. Air from many different sources was encountered, including subtropical air and polluted air from boreal fires in northern Canada. Outflow from thunderstorms contained high $NO_x$ levels indicating that lightning over N. America is an important local source of $NO_x$ and is responsible for part of the $NO_x$ enhancement observed in the North American flight corridor. These measurements and the associated model studies are furthering our understanding of the ozone and trace gas budget in the important UTLS region.

Our understanding of the large-scale circulation of the atmosphere has been improved by comparing a range of models with measurements from a number of sources including $O_3$ from MOZAIC and long-

lived trace gases such as $SF_6$. The results from these and all the transport and mixing studies are resulting in improvements to the model descriptions of transport.

*Arctic vortex*
Observed ozone losses in the vortex were smaller than in previous winters because 1997/98 and 1998/99 were warmer. In 1998/99, models seem to have overestimated the amount of ozone loss compared to the observations. A critical event here is the period in February 1999 when temperatures dropped to PSC (Polar Stratospheric Cloud) existence temperatures but no PSCs were observed. THESEO measurements of chlorine, bromine and nitrogen species are providing a stringent test of model activation in these conditions, with comparisons showing too much activation (and hence too much ozone loss) in models after this event. The most likely reasons for the discrepancy are simplistic microphysics schemes or cold-biased temperatures. This issue of threshold activation is important for mid-latitudes as well as the vortex as regions of threshold temperatures are often found in and around the vortex edge.

The activation process depends critically on the particles which develop as the air cools. One of the great uncertainties has been the chemical composition of these particles. During THESEO, in January 1998, the first measurements of the chemical composition of PSC particles were made on a balloon launched from Kiruna, Sweden. Detailed analysis of the amounts of $HNO3$ and $H2O$ measured in the particles along with optical data indicates that the particles could consist of a supercooled ternary solution (STS) of $HNO_3$, $H_2O$ and $H_2SO_4$, although there are also indications that the particle composition is more complicated.

Modelling studies of denitrification in the Arctic have shown that the observed denitrification can be explained by assuming that relatively large NAT particles can nucleate on ice crystals. This denitrification would result in a larger ozone loss. A colder stratosphere, as might occur as climate changes, could therefore experience larger ozone losses for a given amount of chlorine.

*Tropics and sub-tropics*
For the first time a large number of *in situ* observations were obtained in the tropical upper troposphere and lower stratosphere between the equator and 20°S during the deployment of the M55 Geophysica and DLR Falcon aircraft in February/March 1999 from the Seychelles (5°S). Measurements showed that very thin sub-visible cirrus cloud extended over considerable distances which is being interpreted as consisting of particles made of nitric acid trihydrate, normally observed in polar regions. The existence of such extensive clouds could have important chemical and radiative consequences.

Long-lived tracer and ozone measurements in the tropical lower stratosphere show that most of the air has slowly ascended from the tropical tropopause in isolation from mid-latitude air. In the upper troposphere a weak north-south gradient is observed for tracers with inter-hemispheric differences at the surface, suggesting that air masses from the two hemispheres ascending within the Inter-Tropical Convergence Zone are not completely mixed on entry to the stratosphere.

## 1. BACKGROUND

The overall scientific objective for THESEO (1998/99) has been to increase our understanding of the observed ozone trends over northern mid-latitudes. These reach a peak in the late winter and early spring, but are also apparent in the early winter. Most of the trend in total ozone has come from changes in the low stratosphere at altitudes between 15 and 20 km, the layer which contributes most

to the annual cycle in total ozone. While it is well established that CFCs and other ozone depleting substances are involved, an insufficient understanding of a number of critical issues has limited the ability of the leading computer models to reproduce the observed behaviour. An improved quantitative understanding of the following issues was needed to provide a sound basis for future policy decisions aimed at minimising ozone depletion.

EASOE (1991/92) and SESAME (1994/95), the previous European campaigns, sought to improve understanding of the causes of short-term ozone loss. THESEO concentrated on ozone changes occurring on longer time scales, involving studies of the ozone loss mechanisms operating at different locations and over different timescales, and the connection between them.

The campaign involved a combination of measurements from 35 ground stations, from nearly 40 large, small and long duration balloons, over 1000 ozonesondes and from 5 aircraft deployed from the tropics to the polar regions. Their ability to investigate specific short-duration phenomena was complemented by satellite data and the long term measurements such as those made in the framework of the Network for the Detection of Stratospheric Change (NDSC). Over 400 scientists from more than 20 countries have been involved in the various facets of THESEO.

To maximise the benefits of campaign activities, extensive numerical modelling of the atmosphere was undertaken: meteorological forecasts during the active phase were essential for the planning of operations, and have greatly assisted the interpretation of the measurements. One important aspect of modelling is the use of measurements to improve the descriptions of the tropical stratosphere and the mid-latitude lowermost stratosphere in the chemical transport models.

The research within THESEO has been supported by the European Commission and national agencies. Fifteen THESEO projects have been supported by the Environment and Climate Programme in the EC Research DG. These projects formed an important part of the whole programme on stratospheric ozone and UV-B radiation during that period, which additionally included laboratory-based research into the fundamental principles of stratospheric chemistry, the development of new devices to measure the atmosphere's composition, research into improving atmospheric chemical models, and UV-B field measurements. A detailed description of the scientific aims, the projects and the participating institutions of THESEO can be found in the THESEO planning document (EUR 18177).

In the next section we give an overview of the stratospheric conditions during the campaign. This followed, in section 3, by summaries of important results to date. Finally we describe some on-going advances in research capability which have significantly enhanced the measurements and interpretation during THESEO.

## 2. OVERVIEW

With the wide range of activities that have taken place within THESEO, it is not possible to give a comprehensive summary of all the results in the space available here. Instead, an overall view of THESEO is presented and in the next section preliminary results are summarised in six important areas: Arctic ozone loss; mixing and transport; ozone loss at mid-latitudes; particle composition; halogen chemistry and nitrogen oxides. The findings presented here are based on the most up-to-date analyses available at the time of writing (March 2000). However they are primarily based on the extended abstracts of the results from THESEO which can be found elsewhere in this volume of the proceedings of the Fifth European Workshop on Stratospheric Ozone (which contains nearly 200 extended abstracts), and the reader is referred to them for more details. Results will also be found in the peer-reviewed literature and the planned Second Assessment of European Stratospheric Research

(planned publication in 2001). We thank all scientists and others involved in THESEO who have contributed to the scientific results presented in this report.

The two Arctic winters (1997/98 and 1998/99) covered by THESEO offered the opportunity to extend the series of studies concerning ozone loss in the Arctic lower stratosphere, which started in the late 1980s and continued throughout the 1990s. Since the first large-scale European campaign, EASOE, focussing on the ozone evolution in the Arctic regions, eight Arctic winters have now been well investigated.

The Arctic vortex during the THESEO winters was relatively warm – indeed 1997/98 and 1998/99 were two of the warmest winters in the 1990s and ozone values were much higher than in the previous few years. The interpretation of the measurements made during these two winters should give a better perspective on the ozone loss at high northern latitudes. Previously the most studied winters had, by chance, been the winters in the 1990s which were characterised by low temperatures and low ozone values in the Arctic vortex. These lower ozone values were partly the result of year-to-year changes in the transport of ozone with lower temperatures associated with slower downward motion at higher latitudes. At the same time the low temperatures led to significant chemical ozone losses of up to 50% at some altitudes.

The higher temperatures during the two THESEO winters (in general 1998/99 was warmer than 1997/98) were accompanied by enhanced ozone values and inhibited ozone loss inside the vortex. Chemical ozone loss did take place in the Arctic vortex (see 3.1), but it occurred under threshold conditions (temperatures around those at which fast chemical loss can take place) which had not been extensively studied before in the 1990s. Relatively high ozone values were seen at mid and high latitudes in both winters. Studies of the long term ozone records at Arosa and Hohenpeissenberg have revealed interesting and potentially important links between total ozone trends and changes in tropospheric circulation patterns such as the North Atlantic Oscillation and the Arctic Oscillation. The indices used in the statistical analyses for these phenomena certainly account for a substantial part of the interannual variability at these sites, including the relatively high ozone values in 1998 and 1999. The possible connection of the long term changes in the Arctic Oscillation with climate raises the possibility of this being a mechanism through which climate change will affect stratospheric ozone.

The two THESEO winters were also more dynamically disturbed, with the vortex moving around a great deal. Indeed the first major mid-winter warming since 1990/91 occurred in December 1998, the longest gap between such events in the existing 41 year record (a 4 year gap was the previous record). These disturbed conditions resulted in extensive exchange of air between the vortex and mid-latitudes and a number of specific cases were closely observed from which much is being learnt (3.2). At the same time, good progress has been made on the understanding of the causes of chemical ozone depletion at mid-latitudes (3.3).

The eruption of Mt Pinatubo in 1991 has been a major influence on ozone amounts with the resultant high values of stratospheric aerosol perturbing the chemistry and dynamics. In stratospheric terms, there have been no significant volcanic eruptions since Mt Pinatubo and the stratospheric aerosol loading has now fallen to background values and is similar to those last seen at the end of the 1970s. This reduction in stratospheric aerosol should result in reduced *in situ* loss of ozone over mid-latitudes.

The chemical composition of particles is important in terms of their effects on atmospheric chemistry and in determining the persistence of particles once formed. Important new advances have been made

in measuring and understanding the composition of stratospheric particles in the Arctic and in the tropics (3.4).

From a chemical point of view, the warmer winters and low aerosol and PSC levels have meant that the stratosphere was chemically less disturbed than in other winters. Combination of THESEO measurements with those from previous winters has meant that studies of the important chemical families have now been made under a wide range of conditions (3.5 & 3.6).

## 3 PRELIMINARY RESULTS

### 3.1. Arctic ozone loss

Ozone loss has been studied to differing extents in all the winters in the 1990s, and a good qualitative picture of the involved processes has emerged. Cold temperatures in the stratosphere lead to the presence of cold particles (either PSCs or cold sulphate aerosol) on which fast heterogeneous reactions occur converting inactive chlorine species into active ones. In the presence of sunlight, rapid ozone loss then occurs. Stratospheric models now capture most features of the ozone loss, but there are still periods where there are significant quantitative disagreements and there is a general tendency to calculate less loss than is inferred from measurements. Recent work has therefore concentrated on the quantitative aspects of Arctic ozone loss and has involved a close interplay between the model studies of ozone loss and the empirically determined ozone losses. An important feature of this has been to study the links between dynamics and chemistry as only a correct understanding of the dynamical component allows the precise calculation of the chemical ozone loss. As a result the respective roles of the dynamics and of the chemistry in ozone depletion in the Arctic region and at northern mid-latitudes are now significantly better understood.

The two THESEO winters, 1997/98 and 1998/99, were warm winters when compared to earlier ones. There were only short episodes when temperatures were at or about the PSC existence temperatures in the Northern hemisphere. During the 1997/98 winter, a series of stratospheric warmings resulted in enhanced descent of air and high ozone values at mid and high latitudes. Connected with these, there were periods when the minimum temperatures were low enough for chemical activation to occur and so there were periodic opportunities for ozone loss to occur. In the second winter, temperatures were low early on and PSCs were observed over northern Scandinavia at the start of December 1999. However this was soon followed by the major mid-winter warming and the only other period when minimum temperatures were close to PSC existence temperatures was in early February after which they again rose.

In the 1997/98 and 1998/99 winters, the total ozone losses inferred from the SAOZ network were 20% and 5% respectively. The effect on the total ozone column at the same locations calculated by the two 3D models, SLIMCAT and REPROBUS, was 8-12% in 1997/98 and 5 and 10% in 1998/99. In 1998/99, 25-30% depletions of ozone were calculated by SLIMCAT at around 18km altitude, (compared with slightly larger losses of 30-40% in the previous two winters). Smaller losses were found in 1998/99 at this altitude by the Match technique. Interestingly, these values seem to be higher than the losses deduced from observations and so do not follow the general tendency that the models calculate too little loss. The short periods of low temperatures in the 1997/98 and 1998/99 winters offer a good opportunity to investigate the behaviour of models in marginal temperature conditions (3.5). A particularly interesting event is the cold period in early February 1999, after which over half of that winter's ozone loss in the models is calculated to have occurred.

Research has also concentrated on understanding the interannual variability of ozone loss during the 1990s. The analyses of various stratospheric parameters (minimum temperatures, area of cold temperatures, frequency of stratospheric warmings, etc) from the late 1950s through the 1990s provide a better perspective on the winters of the last decade and an improved characterisation of each particular Arctic winter. Empirically determined Arctic ozone losses from Match and/or the SAOZ network are now available since 1991/92. There is significant interannual variability in the observed loss rates, and the temperature and timing of cold periods are critical factors in determining the ozone loss in a given winter. The warmest winters (1993/94, 1997/98 and 1998/99) are those when there was least loss. Further the winters when temperatures stayed longest around the activation threshold (1994 and 1999) are those where the discrepancies between the models and the empirical estimates of ozone loss are largest.

The weather prediction models used for standard daily weather analyses and forecasts include descriptions of the stratosphere as it is an important factor in producing accurate weather forecasts. At the same time, stratospheric researchers use the meteorological analyses from the weather prediction models in the interpretation of the measurements. For example, the 3D CTM models rely on the temperatures and winds from meteorological analyses – SLIMCAT uses UKMO analyses and REPROBUS uses ECMWF analyses. A particularly important parameter for studies of ozone loss is temperature and it is important that the temperatures in the meteorological models are accurate especially when studying processes which involve temperature thresholds such as chlorine activation. One way in which this has been investigated during THESEO was to make measurements of temperature and other parameters on long duration balloons flown inside the Arctic vortex. These measurements showed significant discrepancies at higher altitudes with the ECMWF temperatures available during the winter of 1999 significantly colder than observed (15 K or more at 10 hPa) while those of UKMO and NCEP were 20 K warmer at 5 hPa. However, these large differences are not apparent in the analyses from the newly available 50-level ECMWF model, which includes assimilation of temperature data from a new satellite instrument (AMSU), where the differences are on average about ±1.4 K at all altitudes covered by the long duration balloon measurements.

### 3.2. Mixing and transport

A good understanding of how the mid-latitude lower stratosphere is linked to other regions of the atmosphere is central to a good understanding of mid-latitude ozone loss. There are two main aspects: (a) the large-scale circulation, the gross features of the movement of air through the stratosphere; and (b) the more localised processes which drive this large scale circulation. These are now considered in turn.

#### 3.2.1 Large scale transport

*In-situ* tracer measurements using balloon-borne whole air samplers were made in winter and spring 1998/99 in order to investigate the extent of mixing between polar vortex air and mid-latitude air. The tracer data are being compared with similar measurements which were carried out in winter and spring 1996/97, when the polar vortex was extraordinarily robust in March and April, and persisted into early May. Inside the Arctic vortex "old" air masses (in terms of mean age and photochemical exposure time) descended from the upper stratosphere/lower mesosphere during winter. After the break-up of the polar vortex, blobs of vortex air can maintain their integrity for up to one or two months and can be advected to lower latitudes, as seen in measurements made at the end of June in 1997 over Gap (Southern France, 44°N).

These measurements also revealed elevated mixing ratios of molecular hydrogen which suggests that this filament comprised fractions of mesospheric air. This finding has an important implication for mean age determinations using the inert tracer sulphur hexafluoride ($SF_6$). $SF_6$ is decomposed by UV

absorption (□□< 125 nm) and electron reactions that become relevant only in the mesosphere. If a significant amount of $SF_6$-depleted air is transported downward into the stratosphere, the mean age determined in stratospheric air samples would be significantly biased towards higher $SF_6$-age values. This is expected to be the reason for the discrepancy between the mean age values derived from $SF_6$ and $CO_2$ in this specific air sample. However, all other air samples of the balloon flights in winter/spring 1996/97 exhibited a good agreement between $SF_6$ and $CO_2$ age values.

For the first time a large number of *in situ* tracer observations were obtained in the tropical upper troposphere and lower stratosphere between the equator and 20°S during the deployment of the M55 Geophysica aircraft in February/March 1999 from the Seychelles (5°S). A newly developed gas chromatograph provided measurements of a variety of long-lived tracers ($N_2O$, F12, F11, H-1211, $SF_6$) along with high resolution $CO_2$ measurements by an infrared analyser. The vertical profiles and tracer correlations with ozone in the lower tropical stratosphere reveal that most of the air sampled has slowly ascended from the tropical tropopause largely, though not completely, in isolation from mid-latitude air. In one flight, however, a recent intrusion of southern mid-latitude air to 11°S at 19 km altitude is apparent. In the upper troposphere a weak north-south gradient is observed for tracers with significant interhemispheric differences at the surface, suggesting that air masses from the two hemispheres ascending within the Inter-Tropical Convergence Zone have not completely mixed by the time they reach the stratosphere.

The performance of different global models in the mid-latitude lower (most) stratosphere has been investigated using measurements. A detailed comparison of 1996 has been carried out with approximately 600 ozone sonde profiles and ozone observations from over 2000 commercial aircraft flight during the MOZAIC project. The models involved are 3D models, which are either climate models or use analysed meteorological fields to calculate the transport. In general, all models provide a reasonable representation of the lower stratosphere. However, the climate models (GCMs) underestimate the build-up of ozone in the lowermost stratosphere during winter and spring, and the models using ECWMF winds (CTMs) overestimate ozone in the layer between 370-425 K during the same seasons. Sensitivity studies reveal that an accurate description of $O_3$ in the 20-50 hPa layer is crucial for the $O_3$ abundance in the lowermost stratosphere.

3.2.2 Links between regions

The relatively high temperatures in the Arctic regions during the 1997/98 and 1998/99 winters were associated with a disturbed vortex, unstable and often displaced from the high latitudes. During the winter and spring in both 1998 and 1999 considerable mixing of air took place between polar and mid-latitude regions. Specific studies within THESEO investigated mixing processes between the different latitude regions and their impact on mid-latitude ozone evolution. In addition, the exchange of tropospheric and stratospheric air near the subtropical jet and the influence on the ozone distribution in this region were investigated. These experimental studies were based on a combination of aircraft measuring in the lower stratosphere, ground-based and ozonesonde measurements made on alert. A good set of case studies during the two years has been collected. New numerical tools, e.g. using nested mesoscale models, were developed in parallel efficiently supporting the analysis of the data.

With an unstable vortex, several events of polar filamentation occurred during the winter. They were observed by the ground based network of ozone lidars and ozonesondes. Six alerts of polar filamentation were sent to the network between mid-December 1998 and March 1999. During the first one, corresponding to the mid-winter major warming, a large fraction of the vortex air (about 50% at 475 K) was being irreversibly mixed with the mid-latitude air. Several polar and sub-tropical filaments were observed between mid-January and the end of April 1999 by the ozone airborne lidars on board the French Mystère 20 and the DLR Falcon. The mixture of polar, mid-latitude and sub-

tropical air over mid-latitudes was also observed in the balloon-borne measurements of tracers such as $H_2O$, $CH_4$ and CFCs in April and early May. The location of the observed filaments was relatively well predicted by high resolution advection models on isentropic surfaces. The comparison of successive cross-sections of the same filament made at short intervals showed no detectable mixing. These results are consistent with current estimates of horizontal diffusion and a mix-down time of 10-20 days.

Ozone measurements from aircraft and ozonesondes have been used to study the exchange of tropospheric and stratospheric air near the subtropical jet. These showed large amounts of ozone (>200 ppb) in thin vertical layers (<1 km) on the equatorial side of the subtropical jet. Conversely layers with low ozone (<200 ppb) transported from the tropics are found in the mid-latitude lowermost stratosphere. Ozone vertical cross-sections measured by an airborne lidar were used in order to initialise and validate a mesoscale transport model and a high resolution advection model. It was shown that both models are able to reproduce the observations and so they can be used to provide reliable estimate of the ozone exchange across the subtropical jet. Statistical analysis of the few ozonesonde stations in the Northern subtropics have also revealed the important role of the Indian summer monsoon.

Detailed distributions of a variety of trace gases in the upper troposphere and lowermost stratosphere over northern mid-latitudes were found during the deployment of the Dutch Cessna Citation. The aircraft flights in July 1998 were carried out over Canada, north of the Great Lakes, over the Atlantic Ocean, and Western Europe. Strong convective activity was present that can transport pollutants into the upper troposphere, partly originating from the boreal fires occurring in northern Canada. More frequently, however, very clean air was encountered in the convective outflow of thunderstorms in the upper troposphere. However this air contained high concentrations of $NO_x$, which can be largely attributed to lightning activity. Lightning over the N. American continent appears to be an important $NO_x$ source that can explain part of the enhanced $NO_x$ levels previously observed in the upper troposphere over the northern Atlantic Ocean. This natural $NO_x$ source provides an important background to which aircraft $NO_x$ emissions are added in the North-Atlantic Flight Corridor.

The lowermost stratosphere was characterised as a mixing layer with enhanced water vapour and tracers with tropospheric origin (such as CO, $N_2O$, $CO_2$, F11, and F12). The thickness of the layer and the slopes of different tracer-tracer correlations within this layer depend on the sample location relative to the jet stream. Southwards at the anti-cyclonic side, the mixing layer is thinner than northwards at the cyclonic side, where the mixing layer extends up to potential temperature levels of approximately 360 K. In addition, the differences in slopes suggest that the air north of the jet stream is predominantly influenced by mid-latitude mixing associated with frontal activity, whereas at the anti-cyclonic side the air originates from the (sub) tropics.

Finally, a comprehensive set of in situ chemical measurements were collected in the lowermost Arctic stratosphere and tropopause region in Jan./Feb. 1997 and 1998 during the POLSTAR 1 and 2 experiments using the DLR Falcon and a Learjet. Compact tracer correlations were observed between $NO_y$ and $O_3$, $NO_y$ and $N_2O$, and $O_3$ and $N_2O$. During both campaigns no signatures of nitrification events were detected in the sub-vortex region. While in POLSTAR 1 the measured tracer correlations compare quite well with results of previous measurements, the inferred $NO_y$-$N_2O$ and $O_3$-$N_2O$ slopes for POLSTAR-2 in 1998 were significantly lower than previously observed in this region. Analyses of air mass trajectories indicate that this is caused by transport of air from lower latitudes to the Arctic sub-vortex region.

### 3.3. Ozone loss at mid-latitudes

The causes of mid-latitude ozone decline remain controversial and their study was a major focus within THESEO. It has been argued by some that much of the loss occurs *in situ* while others have suggested that polar processes have a very significant influence on the decline of ozone in mid-latitudes. A number of studies within THESEO, both observational and modelling, are making an important contribution to the debate.

A major effort within THESEO has been to develop empirical techniques capable of measuring chemically induced changes in ozone over mid-latitudes. These changes are hard to detect as they usually occur at much slower rates than in the Arctic and distinguishing the effects of chemistry and dynamics is trickier. However promising starts have been made with a couple of approaches, whose preliminary results show that chemical ozone changes occurred in air outside the Arctic vortex and which are distinguishable from processes occurring inside the vortex.

First, the cumulative column loss at latitudes south of 60°N, inferred using a combination of SAOZ measurements and the passive ozone in the Reprobus model, is found to vary between 5 (in 1998/99) and 15% in the last 6 winters. However a number of factors still need to be examined and accounted for, not least the fact that there is $NO_x$ catalysed chemical destruction of ozone occurring at mid-latitudes which would be included in the empirical estimates of ozone loss but should clearly not be ascribed to halogen chemistry. Second, preliminary analysis of the Match experiment indicates that ozone loss occurs faster at lower temperatures. In warmer air, no ozone loss can be detected within the uncertainties of the approach. This is in qualitative agreement with current chemical understanding and highlights the importance of the region near the vortex edge where low temperatures are most likely to occur outside the vortex. When confirmed these studies will represent the first direct observational evidence of *in situ* chemical ozone changes outside the vortex.

Chemical transport models have been used to examine the connection between polar and mid-latitude loss. The winter of 1998/99, the second THESEO winter, has proved to be especially interesting. Temperatures in the lower stratosphere were higher than average. There was, however, some activation of chlorine inside the polar vortex although outside the vortex synoptic scale temperatures were too high for activation. There were major dynamical disturbances to the vortex in mid-December and in February. Model calculations show that there was considerable exchange of air between the polar vortex and middle latitudes associated with these events. North of 40°N equivalent latitude, the partial column loss between 350 and 670 K is calculated by SLIMCAT to have been 34 DU in 1998/99, similar to the values in 1995/96 of 43 DU, in 1996/97 of 28 DU and in 1997/98 of 35 DU. There was only a relatively small amount of chlorine-induced loss but this manifested itself as ozone loss in the model throughout middle and high latitudes. Model loss rates did not show a large latitudinal gradient. Clearly, in this winter at least, there was a strong connection between ozone loss in middle and high latitudes. The accuracy of the model estimates, which seem in this winter to be higher than those derived empirically, will depend on the amount of activation that actually occurred in February 1999 and it is important to make a thorough comparison between the observed and modelled activation during this second period of cold temperatures (3.5).

Interesting comparisons can be made with the modelled ozone losses in earlier winters, including the first THESEO winter, 1997/98. For example, the vortex of 1996/97 was stable and long-lasting with low temperatures again confined inside the vortex. Model calculations indicate that, while there was substantial ozone loss inside the vortex, there was only a modest decline of ozone outside the vortex. This is in contrast to the model results for the recent, more dynamically disturbed winters which show a larger loss outside the vortex, and less contrast with the in-vortex depletion. If these results about the connection between polar and mid-latitude ozone loss are generally true, then a cold, stable vortex

favours large polar loss with little connection with middle latitudes until the final vortex breakdown, but in more disturbed winters polar loss, and polar processing, contribute to mid-latitude decline.

### 3.4. Particle composition

The first measurements of the chemical composition of Polar Stratospheric Cloud (PSC) particles were made during THESEO. These measurements were obtained using a newly developed aerodynamic focusing lens in combination with a mass spectrometer which was flown on a balloon launched from Esrange, Sweden, in January 1998. The particles are separated from the ambient gas so that the mass spectrometer can provide an analysis of the nitric acid and water content in the particles. On board the gondola was also a backscatter sonde which measured the cloud particle backscatter ratios at two wavelengths. These real-time observations of backscatter were used with great success to manoeuvre the balloon through three passages of PSC layers during the flight.

Whenever the backscatter signals increased above the stratospheric background levels, indicating PSC particle growth, strong $HNO_3$ and $H_2O$ signals were measured, confirming earlier observations that PSC particles are composed of these substances. The molar ratios between $H_2O$ and $HNO_3$ were well above 10, indicating that the particles were not composed of nitric acid hydrates, but rather pointing to a liquid supercooled ternary solution (STS) composition. An analysis of the chemical and optical measurements, using microphysical and optical models, reveal however that the composition of the particles could be more complicated. New experiments are planned for the THESEO-2000 campaign with additional instrumentation on the PSC-analysis gondolas to obtain information about the size distribution and physical phase of the particles together with the water vapour concentration in the gas phase.

PSC particles make possible the chemical activation of chlorine species into ozone depleting radicals in the polar lower stratosphere. These heterogeneous reactions depend on the chemical composition and physical phase of the particles. In addition, solid PSC particles are responsible for denitrification in the lower stratosphere, i.e. the irreversible downward transport of reactive nitrogen which would otherwise limit the ozone depletion from activated chlorine. Microphysical details about the solid PSC formation are unknown. Denitrification has been observed occasionally in the Arctic polar stratosphere in contrast to Antarctica where this phenomenon occurs regularly every winter. It is currently an open scientific question if predicted lower stratospheric temperatures, arising from increased concentrations of greenhouse gases in the atmosphere, could lead to more widespread PSC formation and denitrification and thereby stronger ozone depletion in the Arctic in a future climate.

It has been suggested that cirrus clouds provide an important sink for $HNO_3$ in the upper troposphere and near the tropopause. During the POLSTAR aircraft campaigns based from Kiruna $NO_y$ uptake by cirrus clouds were investigated for different atmospheric conditions. Gas- and particle-phase $NO_y$, total $HNO_3$, total $H_2O$, together with particle concentration and size were measured from the DLR Falcon during flights through various cirrus clouds in the vicinity of the tropopause at latitudes between 60°N and 70°N. In the cirrus clouds only a low uptake of 5-15 pptv $NO_y$ (as $HNO_3$) onto ice particles was seen. The total available abundance of $HNO_3$ was larger by factors of 6 to 50. A possible explanation for the limited $NO_y$ uptake by cirrus clouds include the presence of other, competitive adsorbers on the surface of ice particles.

Measurements of the physical and chemical composition of particles were also made in the tropics within APE-THESEO. Innovative flight planning meant that satellite cloud data and real-time aerosol lidar data from the DLR Falcon were used to guide the Geophysica. Thus the high flying aircraft was able to penetrate the thunder cloud anvils so the *in situ* trace gas and particle measurements were made which have been used to complement the remote measurements from the lidar. Measurements

from both aircraft showed the existence of very thin sub-visible cirrus which extended over considerable distances. This was something of a puzzle as the observed clouds existed across too large a temperature range for the cirrus to consist of ice particles. Preliminary analyses of the measurements from a new instrument indicate the presence of $HNO_3$ in the condensed phase. Model calculations based on measurements from several instruments and on laboratory data indicate that the particles are made of nitric acid trihydrate.

### 3.5. Halogen chemistry

Bromine and chlorine compounds are involved in the depletion of ozone at mid-latitudes. Studies within THESEO have investigated the stratospheric budgets and chemistry of the various species involved so that their chemical influence on ozone can be better quantified.

#### 3.5.1 Bromine compounds

The budget of stratospheric bromine has been investigated using measurements of organic bromine species found in whole air samples (organic bromine method), and by using a combination of BrO measurements and models to infer the inorganic bromine partitioning (inorganic bromine method).

Gaseous organic precursors to $Br_y$ have been measured in whole air samples from the TRIPLE gondola flights. The most complete inventory of organobromine gases was obtained during a flight from Kiruna on 6 February 1999. From the corresponding tropospheric concentrations, the amount of $Br_y$ released by each precursor was calculated at each altitude level. (Adjustments for atmospheric growth were based on dating of the stratospheric air using $SF_6$ and $C_2F_6$ as tracers.) Twelve separate organobromine species were measured and the total estimated $Br_y$ determined in this way was 18.4 ± 1.8 ppt above 20 km (age of air 4–5 years).

Methyl bromide ($CH_3Br$) is by far the largest contributor to $Br_y$ at all altitudes, contributing about 46% of the $Br_y$. The halons (H-1301, H-1211, H-2402, and H-1202) together account for about 27% above 20 km, and the naturally released, short-lived bromoalkanes contribute about 13%. Among them the most important species is bromoform ($CHBr_3$) although the exact amount is critically dependent on the assumed entry level of this short-lived species into the stratosphere. Aircraft and balloon measurements currently indicate at least 0.5 pptv of bromoform (1.5 pptv Br) being transported to the stratosphere. Above 20 km this amounts to about 8% of total $Br_y$, but the contribution may be proportionately larger in the lower stratosphere where the contribution from the longer lived species is much less. Bromoform was closely followed by dibromomethane in terms of contribution to $Br_y$. The contribution from all other species combined amounted to only 2%.

The measurement of total organic bromine in whole air samples has been corroborated by the inorganic bromine method applied to the BrO measurements made by LPMA/DOAS on a balloon flight at Kiruna on 10 February 1999. Total stratospheric bromine content is estimated to be 21.5 ± 3 ppt (in 6 year old stratospheric air), in reasonable agreement with the organic bromine budget. The somewhat larger total bromine inferred from the inorganic bromine method indicates the possibility of an inorganic influx of 3.1 (-2.9/+3.1) ppt from the troposphere into the stratosphere. This conclusion is also supported by recent tropospheric BrO profile measurements showing up to 1 ppt of tropospheric BrO, and about 1 ppt of Br found in the aerosol of the upper troposphere. Although supposedly variable, the inorganic bromine influx into the stratosphere is estimated to contribute 14% to the stratospheric bromine loading. In conclusion, the man-made bromine species contributed 51-58% to the total stratospheric bromine loading of 20 ppt in early 1999.

The consistency of the stratospheric bromine content and chemistry at mid-latitudes has been investigated using balloon-borne BrO observations. For mid-latitude autumn in 1996, excellent

agreement is found between the measured and modelled BrO assuming 20 ± 2.5 ppt of lower stratospheric bromine in the model. This value is slightly less than for the 1999 bromine inorganic budget analysis, but it consistently reveals that the increasing tropospheric halons (about 1 ppt) concentrations have indeed augmented the stratospheric bromine since 1996.

Bromine monoxide has been extensively monitored during THESEO by means of UV-visible spectrometers operated from the ground, balloon and satellite (ERS-2/GOME). Altogether these observations form a data set of unprecedented completeness and have been used to test our understanding of the stratospheric bromine chemistry at various latitudes, seasons, and time of the day. Comparisons with calculations by the SLIMCAT 3D CTM reveal a good overall degree of agreement between the modelled and observed stratospheric BrO amounts and variability, after allowing for a small amount of BrO in the troposphere. For example, the latitudinal and seasonal variability of BrO columns and profiles are well reproduced. SAOZ balloon measurements have revealed that bromine is always activated (60% in BrO during daytime) at all latitudes and seasons. Most of the column changes in BrO previously reported from ground or satellite observations are therefore due to transport (vertical displacement of the stratosphere) and not to chemistry. Photochemically, in order for the models to reproduce the BrO observations, they must simultaneously reproduce the observed behaviour in the $NO_x/NO_y$ and $ClO_x/Cl_y$ ratio. In general however the variability is well reproduced by 3D CTM models, as are the signatures of chlorine activation occasionally observed in twilight BrO columns and explained in the model by the formation of the photolabile BrCl.

Our overall understanding of current bromine sources and chemistry gives us confidence that current models can be used for an assessment of the role of bromine in mid-latitude ozone loss. The increase in the estimate of inorganic Br to about 20 ppt, implies a greater importance for bromine in chemical ozone depletion than previously thought. Some discrepancies between model and observations still exist which require further study, e.g. the behaviour of morning to evening column amounts ratios which are not well reproduced at all ground-based stations, and the partitioning between bromine species is controlled by $NO_2$ which is largely underestimated by the models in the lower stratosphere (3.6).

3.5.2 Chlorine activation

There were two periods of activation in the 1998/99 winter: early December and early February. Otherwise the stratospheric temperatures were too high for activation to occur. The ozone loss rates calculated by models are apparently higher than those deduced from observations and so it is important to examine all the available chemical measurements carefully and to compare them with the modelled chemical amounts. In particular it is important to determine how good and how quantitative our understanding of activation in threshold conditions is. Four aspects of this issue are discussed here: daytime ClO amounts; the interpretation of OClO observations; the ClO and BrO decay at sunset; and activation in the lowermost stratosphere.

The vortex remained activated for some time after the December event, even through the major mid-winter warming. The $ClO_x$ activated in early December decayed through December and January as a result of the low amounts of sunlight and consequently low $NO_2$ production. Slightly enhanced ClO amounts (200-300 pptv measured by ASUR) were observed in mid-January which had decayed slightly (<250 pptv) by early February. On 27 January, a broad peak in $ClONO_2$ vertical distribution was observed by MIPAS-B, consistent with the prior conversion of ClO to $ClONO_2$ through reaction with $NO_2$. However SLIMCAT calculated ClO at the beginning of February as high as 1000pptv (same day, same place as ASUR measured 250 pptv). The period of low temperatures in early February is important as much of the loss calculated in SLIMCAT occurred after this time, so that it is

important to ascertain how much activation took place. No PSCs were observed at Kiruna arou this time, even though local temperatures were 1-2 K below PSC existence temperatures. Groun based measurements at Ny Ålesund showed no ClO activation during February, again in contradicti to SLIMCAT. At Harestua, Norway, closer to the area of cold temperatures, only slightly elevat ClO was observed and much less HCl removal was observed than calculated. At the same time OC amounts measured inside the polar vortex were significantly smaller than calculated. The comparisons indicate that SLIMCAT activates too readily, either because of a simplistic activati scheme or because of a cold bias in the temperatures used.

Several measurements of OClO and related species made in mid-February. On 10 February, lar amounts of OClO were measured by LPMA-DOAS although prior to the balloon flight stratospher temperatures were above the PSC existence temperature. This observation can be interpreted resulting from the efficient hydrolysis of $BrONO_2$ on cold stratospheric aerosol. Night-tim measurements of the vertical distribution of OClO and other species were made by AMON on February again inside the vortex. The OClO mixing ratio was found to be relatively high (50 to pptv). Calculations using the MiPLaSMO lagrangian model show that these values can be reached night in presence of a very small activation, if the $NO_2$ mixing ratio is low. At low altitudes (50- hPa) low $NO_2$ amounts were measured, and so the model and the measurements of $NO_2$ and OClO a in agreement. However at higher altitudes, the computed OClO mixing ratio is lower than observed the model is constrained to fit the measured $NO_2$. Conversely if the model is constrained to fit OCl it cannot reproduce the relatively large measured $NO_2$ mixing ratio. This result shows that chlorin nitrogen interaction is not yet fully understood in these conditions.

Observed and modelled ozone loss rates agree poorly at high solar zenith angles. Resolution of th problem could explain a significant part of the overall discrepancy between models and measur ozone losses, particularly in winter. Consequently an *in situ* ClO/BrO instrument was flown as part the TRIPLE balloon payload on 6 February 1999 to investigate the sunset variation of ClO and Br The sunset variation of the radicals in the solar zenith angle regime from 88° to 94° was measur while floating in a polar vortex air mass at around 63 hPa (18 km, 460 K). The measurements we compared to the output of chemical box model runs which were initialised from simultaneo measurements and using known correlations. While the BrO variation at high solar zenith angles well reproduced by the model the ClO mixing ratios at solar zenith angles higher than 86° a markedly underestimated. Possible sources of the observed discrepancy are the overestimation twilight $NO_2$ or an underestimation of the twilight sources of ClO due to problems with the mod photolysis rates at high solar zenith angles. If the current disagreement can be established this wou explain part of the observed underestimation of the modelled ozone loss when compared measurements.

Indirect evidence of chlorine activation has been obtained from aircraft measurements in t lowermost stratosphere by the observation of anomalously enhanced $CO/C_2H_6$ ratios. T measurements also confirmed the presence of subvisible cirrus above the tropopause, suggesting th local chlorine activation has taken place on cirrus crystals. Furthermore, a chemistry box-model and three-dimensional chemistry-transport model were used to investigate the photochemical effects enhanced ice particle surface areas in the lowermost stratosphere. It appears that ice particles in t lowermost stratosphere can cause chlorine activation and chemical $O_3$ loss during most of the yea not only limited to the cold seasons (winter and early spring).

These studies show that there are still a number of open questions concerning the chemistry chlorine radicals and certainly the coupling between the reactive chlorine, bromine and nitroge families. Further the treatment of activation in 3D models seems to be problematic in threshol

conditions as the SLIMCAT and REPROBUS models seem to have overestimated the ozone loss which occurred after the cold period in February 1999 even though they use different temperature analyses. (SLIMCAT uses UKMO analyses; REPROBUS uses ECMWF analyses.)

### 3.6. Nitrogen oxides

A number of data sets on the nitrogen partitioning are now available from different geophysical situations at mid and high latitudes. These are being used to check the validity of the nitrogen chemistry in the models and - together with simultaneous $N_2O$ measurements - potential denitrification events. These are now considered in turn.

The observed $NO_x$ concentrations in the lower stratosphere are still significantly larger than those modelled. Recent revisions of laboratory data of chemical reaction rates for the reactions of $NO_2$ and $HNO_3$ with OH have reduced the differences in modelled and measured concentrations as well as in the delay in the $NO_x$ recovery at spring. However significant discrepancies remain which in terms of the $NO_x/NO_y$ ratio appear to be dependent on the solar zenith angle and on the amount of $NO_x$. Models still fail to capture the presence of $NO_x$ at spring in the lowermost stratosphere as in the model there is fast conversion of $HNO_3$ on cold aerosol in contrast to observations. Since the discrepancy is similar every year in the long series of profiles started in 1992 when the aerosol loading was 10 times larger, the likely explanation would be in the $NO_x$ gas phase chemistry at lower temperatures which should be re-examined. This issue needs further attention since the amount of $NO_x$ controls the deactivation of chlorine. Moreover, model-observation comparisons show that either the chlorine chemistry or the $NO_y$ chemistry can be brought in consistency with model outputs, but not both families simultaneously.

In order to study the effect of the photolysis rates at high solar zenith angles, the photolysis rates for $NO_2$ measured from aboard the LPMA/DOAS gondola were compared with two radiative transfer models. The good agreement between the measurements and the models allows us to rule out errors in the adopted $JNO_2$ photolysis rates to be a major cause for the large discrepancy found in measured and modelled stratospheric $NO_2$ in high latitudes at large SZAs (pm and am).

Profiles of total reactive nitrogen ($NO_y$) summed from the individual compounds of the nitrogen family and its correlation with the simultaneously measured source gas $N_2O$ have been used to study denitrification from balloon flights in different winters. Traditionally, the $NO_y/N_2O$ ratio has been used to diagnose denitrification. However if mixing processes are not considered, this method can overestimate or even 'produce' denitrification. On the basis of observed $NO_y/N_2O$ and $CH_4/N_2O$ correlations the uncertainty of the denitrification signal derived solely from the $NO_y$-$N_2O$ correlation has been investigated. The interannual variability of denitrification can be seen in 3 MIPAS-B flights: a strong denitrification appeared in winter 1995; only a weak unambiguous signal was found in March 1997; and no sign of denitrification in January 1999.

Arctic ozone loss in a given winter is greater if denitrification occurs. This is one of uncertainties affecting ozone loss calculations as, to date, 3-D chemical transport models have not been able to reproduce the observed $NO_y$ redistribution. Recently a quasi 1-D microphysical model was able to simulate the denitrification observed by MIPAS-B in 1995 during SESAME by suggesting that the sedimentation is predominantly caused by relatively large nitric acid trihydrate particles with small number densities which have nucleated on ice particles. In a colder stratosphere, as predicted by current climate models, the reduced ozone loss that would result from the expected reduction in chlorine and bromine levels would be offset by the additional periods of low temperatures and the enhanced denitrification associated with them. Efforts to implement parameterisations of sedimentation into 3-D models have recently shown promising results. Denitrification events have to

be captured well in 3-D models in order to calculate the effect of the greenhouse-denitrification-ozon loss coupling correctly.

## 4. CONTINUED DEVELOPMENT OF RESEARCH CAPABILITY

The experience of THESEO has emphasised the advances in expertise and research tools that hav taken place during the 1990s. It is easy to take these advances for granted, but much successfi research is achieved by sustaining developments over long periods. These advances has been realise in a number of ways described below, but possibly the most important is the development of community of researchers who understand the advantages of collaborating on a pan-European basi One group worth particular mention are the many young scientists who have greatly benefited fro the additional breadth of scientific training available through working with leading scientists fro other countries in European projects.

### *a) Improving capability of ground-based networks*

The capability of the European network of ground-based instruments (uv-vis, FTIR, lidars, microwav radiometers and UVB instruments) has improved significantly in recent years and so the network ha provided a validated and consistent data set of stratospheric species and a continuous monitoring o the chemical evolution of the stratosphere in the Northern hemisphere. There are two main reason First the operators are now experienced and can produce near real-time data of some critical chemica species (e.g. aerosols, HCl, OClO and BrO) which can be used for planning later activities. Second many instruments and their algorithms have been technically improved so that additional informatio on vertical profiles is available and the quality of the measurements improved. These changes hav noticeably improved the scientific usefulness of the ground-based observations at existing high-latitud and mid-latitude stations in Europe particularly for the efficient support of other field activities durin large campaigns.

### *b) Proven techniques for empirical Arctic $O_3$ loss determination*

During the 1990s a number of techniques based on measurements have been developed to estimate th amount of chemical ozone loss in a given Arctic winter. The results have been used to build up picture of the conditions (temperature, sunlight, etc.) under which ozone loss occurs in the Arctic an to test the accuracy of the model calculations of ozone loss rates under a wide variety of conditions The smaller ozone losses (at the limit of detection) reported in 1999 when only limited activatio occurred gives confidence in these techniques. The discrepancies revealed are being used to identif new or incorrectly described processes and so improve the model description of Arctic ozone loss. I is these empirical techniques which are being developed so that chemical ozone changes can b identified at mid-latitudes.

### *c) Utility of 3D chemical transport models*

Three dimensional chemical transport models driven by real wind fields from weather predictio models have developed enormously in the last ten years, with European scientists in the vanguard o research in this area. These sophisticated models came of age during SESAME, and during THESE they have been extensively compared with field measurements, with studies varying from detailed photochemical comparisons of the diurnal variation at sunset to comparisons with long time series o measurements made by ground-based instruments. The 3D CTMs provide global and continuous results (geographical scale, time). During the THESEO campaign, they showed their ability and thei utility to investigate in a global scale various chemical and dynamical processes such as $O_3$ loss, mixing, filamentation, and transport between the different latitude regions. Meso-scale 3D model and high resolution advection models have also proven valuable because of their high vertical resolution which makes possible reliable estimate of air mass transport across dynamical boundaries.

*d) Improvements in balloon-borne instrumentation.*

A traditional strength of European science has been in measurements made using balloons. These have typically been large payloads (hundreds of kg) which are capable of measuring many species of interest simultaneously. These instruments have been improved in terms of the number of species they measure and in the precision of the observations they make. Detailed comparisons between instruments (e.g. for the SPARC water vapour report) and with models have been successfully made. Recently a number of small balloon instruments (3-40 kg) have been developed which are relatively easy to deploy and which measure a smaller number of species. During THESEO these instruments have been used quite extensively on small and long duration balloons, and occasionally as guest instruments on some of the larger payloads. These new instruments represent a significant addition to the European observing capability.

*e) Available, instrumented aircraft for 10-14 km*

Several aircraft exist which fly in the 10-14 km altitude range have had considerable use in recent years. There are two main types of payload: remote-sensing payloads such as a lidar which can measure vertical profiles above the plane; and *in situ*, multi-instrumented payloads capable of measuring many species simultaneously. During THESEO the former were used in studies of PSCs, filamentation and chlorine activation at high latitudes and the latter were used to study the transport of air in the lower stratosphere and upper troposphere with the large numbers of measured chemical species being used to identify a wide range of air masses.

*f) Potential for Geophysica as a high-flying platform*

At higher altitudes (up to 20 km) the Geophysica proved itself to be a reliable high-flying platform during its APE-THESEO deployment in the Indian Ocean in February-March 1999. For example, in conjunction with the lidar on the DLR Falcon, it was guided into the core of convective cloud systems so that measurements could be made of chemicals and particles in air which had been recently lifted up from ground level. A similarly flexible approach to flight planning was shown during the APE-GAIA campaign in the Antarctic in September-October 1999 when it was able to fly through mountain-wave PSCs over the Antarctic peninsula.