

Report of the 9th Session of the SPARC Scientific Steering Group

Honolulu, Hawaii, USA, December 4-7, 2001

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This ninth session of the SPARC Scientific Steering Group (SSG) was held in Honolulu, Hawaii, thanks to the kind invitation of Kevin Hamilton. The session was opened by the co-chairs M. Geller and A. O'Neill who both pointed out in their preliminary comments the fact that SPARC was at a turning point and should move forward with the integration of activities as discussed at last year's SSG. They mentioned the present situation with IGAC in preparing a new phase and the close relationship that might develop between the two projects during this phase change. A. O'Neill remarked that the chemistry/climate issue was not yet prominent in either the SPARC or IGAC programs, whereas the subject was going to be a priority issue in the following years.

After the logistical and practical information given by K. Hamilton, the meeting started by a review of the



Participants of the SSG in Honolulu.

First row: K. Hamilton, M. Geller, A. O'Neill, M.-L. Chanin

Second row: M. Shiotani, V. Ramaswamy, P. Udelhofen, R. Vincent, Ph. deCola, V. Yushkov, J. Gille.

Third row: T. Peter, W. Randel, L. Thomasson, M. Baldwin, A.R. Ravishankara, C. Michaut, U. Schmidt, M. Proffitt, R. Newson, D. Karoly, Y. Koshelkov, T. Shepherd, S. Pawson.



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main events which took place in the last year, in particular the meeting of the WCRP Joint Scientific Committee (JSC), that took place in Boulder, USA, in March 2001. During the JSC the SPARC co-chairs, M.-L. Chanin and M. Geller made presentations of the SPARC programme, including the results from the SSG meeting in Buenos Aires, some of the discussion on SPARC future and a summary of recent results relating to solar effects on climate.

According to R. Newson, the JSC thought SPARC was going well. He noted that there were good working relationships between GRIPS and the WGNE, and that SPARC was playing an important role to achieve a closer cooperation between WCRP and IGBP. Therefore the JSC did endorse the idea of a joint SPARC/IGAC Planning Group for the future. The JSC also asked S. Solomon and G. Brasseur to initiate discussions meant to look into a future chemistry/climate project. They also thought it would be good to look into the possibility of an AO/NAO workshop that might involve ACSYS/CLIC, CLIVAR, and SPARC. The JSC did support the SPARC suggestion of a joint SCOSTEP/SPARC Working Group to assess upper atmosphere temperature trends. They did not support the idea of a joint WCRP/SCOSTEP Working Group to assess the role of the sun in climate change. They did, however, ask GEWEX to look into the cloud data sets that have been used recently to indicate solar effects on cloud cover and to comment on the use of those data sets to conclude the existence of solar effects on cloud cover.

Modelling stratospheric effects on climate

Intercomparison of stratospheric models

The progress report of GRIPS ("GCM Reality Intercomparison Project for SPARC") was given by S. Pawson. After the important activity leading to the publications of the first GRIPS results in 2000 (BAMS and JGR), it was recognized that GRIPS had grown considerably both in the number of groups involved and the range of tasks being tackled. A complete discussion of the ongoing activities took place during the GRIPS workshop, which was held in March 2001 in Hamburg, Germany. The state of achievement varies for the different tasks, while

some tasks of phase 1 (model intercomparison) are completed (basic climatology, travelling waves/tides, spatial spectra), others are ongoing, and some are stagnating. Within task 2 (parameterisations/impacts), the test of radiation schemes is in progress and will facilitate moving forward with the gravity wave (GW) drag parameterisation, while the study of model response to mesospheric drag is completed. Activities related to phase 3 (forcing by changes in aerosols, ozone, solar radiation, CO₂), are being tackled by a few groups, some of them in connection with the European projects SOLICE and EuroSPICE. One of the main challenges of GRIPS this year should be to address the reasons for the divergence between different predictions of evolution of the Arctic, as a contribution to the upcoming WMO-UNEP Ozone Assessment.

It was agreed that the development of phase 3 should be discussed in relationship with the integration of models/data activities within SPARC, as well as the integration of the troposphere/stratosphere coupling issue. The primary goal of GRIPS is to ensure that the coupled models have a good stratosphere model, which implies a close cooperation with WGCM.

The project for 2002 is to hold a new workshop in March 13-17 at Tsukuba, Japan, to wrap up phase 1 (which should be near completion at that time) and to review phase 2 with the aim to complete it in 2003.

Following an invitation by the co-chairs, S. Yoden made a presentation of the Japanese Frontier Project. One of his purposes was to investigate a possible cooperation between SPARC and the Earth Simulator modelling activities, for example on the atmospheric chemistry/climate issue. It was suggested that a presentation on this subject should be made at the next JSC meeting and at the future GRIPS workshop to develop the ideas further.

Stratospheric reference climatology

W. Randel presented the recent inputs into the technical report "SPARC Intercomparison of Middle Atmosphere Climatologies" to be published as a SPARC Report in mid 2002. They mainly consist of the inclusion of rocketsonde and lidar data into the data intercomparisons, which allowed the extension of the climatology of temperature and winds up to the middle mesosphere. He presented their comparison with global satellite data and

indicated the difficulties due to the scarcity of sites and the non-simultaneity of the data sets, most of the rocket series having been discontinued. A group meeting took place in Boulder in September 2001 to complete the report and in particular to identify the biases in each data set, to identify the quantities, which have high uncertainty, and to discuss the strategies for comparison with models. The question was discussed whether or not to include ERA40 from ECMWF into the data intercomparison with the consequence of postponing the publication of the report. It was decided to try to finish the report before spring, and include the ERA40 data only if available.

SPARC data assimilation

The new working group on SPARC data assimilation has now been constituted under the acting chairmanship of A. O'Neill, who presented the project. The goal of the project is to exploit the technique of data assimilation to provide quality-controlled, internally consistent global data sets of the dynamical and chemical state of the stratosphere (as well as, where possible, the upper troposphere and mesosphere). The data sets will be targeted at SPARC-related activities to understand chemical-climate interactions. The initial focus will be on exploitation of research satellite data from the ENVISAT and EOS AURA satellites. Additional outputs from the project will be error statistics relevant to utilization/validation of instruments and to evaluation of numerical models.

Specific activities will include:

- Comparison of global data sets produced by participating groups,
- Development of documentation at the SPARC Data Centre on data production methods and data quality,
- Workshops to improve data assimilation methodology for the stratosphere (e.g. to assimilate new variables such as aerosols amounts),
- Liaison with WGNE,
- The project will also produce reports on specific aspects of the data sets of topical interest (e.g. stratospheric water vapour and its evolution).

The SPARC working group will foster collaboration and information exchange. Its first meeting took place at the AGU, held in San Francisco the week following the SSG. A working group meeting, to be held at the Data Assimilation Office, Goddard Space Flight Center, is being arranged for June 2002.

Long-term changes in the stratosphere

Stratospheric temperature trends

The summary of the first phase, which covered the period 1979-1994, was published in *Reviews of Geophysics* in February 2001, and the results were used in IPCC/TAR 2001. The full account of the work is also in preparation as a SPARC Report to be edited by NOAA and SPARC in 2002.

V. Ramaswamy presented the update of the annual-mean trend for the period 1979-2000 (see the report in this issue). It confirmed nicely the conclusions published earlier of a general cooling of the stratosphere, but the significance has been much improved. These results will be a contribution to WMO-UNEP Report 2002. He also mentioned new corrections in homogeneities in radiosonde records and new trend results obtained from rocket data. The more recent comparisons between the observed trend and the simulation by models taking into account updated knowledge of the trends in species (ozone and CO_2) changes indicate a reasonable agreement up to 0.5 hPa. The priority should now be to look at the variability both observed and simulated and its implications for trends.

The SPARC/SCOSTEP project of assessing the mesospheric trends was discussed during the IAGA workshop held in Prague in July 2001, which was co-sponsored by SPARC (see report in this issue) and its output was commented by V. Ramaswamy and M.-L. Chanin who attended the workshop, as well as by M. Geller. They greeted with pleasure the proposal by F.-J. Lübken to organise the next workshop on this issue to be held in Kühlungsborn, Germany in May 2002, and the SSG provided suggestions of a few extra names of participants and offered the experience gained in the 3 successive assessments led by SPARC, to ensure that such a work lead soon to a thorough assessment of the mesospheric temperature trends. M. Baldwin agreed to be the SPARC Chair in this working group.

Understanding ozone trends

N. Harris, unable to attend, had sent a report on the activities of the group during the year. It was mainly related to the SPARC/IOC workshop on Understanding Ozone Trends which was held at the University of Maryland, March 7-9, 2001, in preparation for the WMO-UNEP Ozone Assessment 2002.

The main aims of the workshop were to identify the major current issues concerning ozone trends, to improve quantification of the contributions and uncertainties of the chemical and dynamical mechanisms to observed ozone trends, particularly at mid-latitudes and to identify how to assess the consistency of these proposed contributions. The main emphasis was on the dynamical influence on the ozone trends. W. Randel, lead author of Chapter 4 - Global Ozone, presented the current state of the chapter. T. Shepherd and N. Harris are jointly preparing the chapter on the influence of dynamics on the 'observed' trend. T. Shepherd commented on the difficulty to identify separate dynamical forcings in terms of an ozone response. They decided to organise a comparison of the statistical analyses including dynamical factors so that they can be compared for certain baseline periods and it was suggested that this comparison be considered as a SPARC activity.

To move forward on the issue of Arctic ozone, a workshop on Arctic Ozone Loss will be held at the Alfred Wegener Institute in Potsdam, Germany from March 4-6, 2002.

T. Peter presented the European report on THESEO entitled "European Research in the Stratosphere 1996-2000" (see review in this issue). He also provided news about the Geophysica Project for which 5 proposals have been accepted for support by the European Commission, including TROCCINOX on tropical deep convection and its influence of NO_x , and EUPLEX dealing with PSCs and Lee Wave detection. This was seen as very encouraging for SPARC related activities and a letter indicating this will be sent to the EC by the co-chairs.

Stratospheric and upper tropospheric water vapour

The Water Vapour Assessment, SPARC Report No 2, 2000 (and WCRP-113, and WMO-TD-No 1043), which investigated the concentration, distribution, variability and trends of water vapour in the stratosphere and upper troposphere, has been very well received by the community. 400 copies have been distributed and more copies are requested regularly at the SPARC Office, as well as many requests to use figures from the report. The results have been presented widely during different meetings and they are now being used as background material by the community. SPRINGER has offered to publish it as a book and the question is under study.

SPARC Aerosol Assessment

This new project entitled "Assessment of Stratospheric Aerosol Properties" (ASAP) is being led by L. Thomason and T. Peter. Its status was presented at the SSG by L. Thomason.

It is recognized that aerosol in the upper troposphere/lower stratosphere (UT/LS) can have a significant impact on climate through radiative effects and on stratospheric chemistry, particularly impacting ozone. The magnitude of these effects is also highly variable and, as a result, can complicate isolating human-derived changes in the stratosphere. Accounting for aerosol effects properly is a key component of modelling climate/chemistry effects properly.

The goal of this new SPARC initiative is to evaluate the scientific understanding of UT/LS aerosol and aerosol measurements. The assessment was initiated with a workshop in Paris, France on November 4-6, 2001. The questions to be addressed in this work and how they will be tackled are described in the report from this workshop published in this issue. The goal is to complete the assessment in less than 2 years. The results will be published as a SPARC Report and the data will be available at the SPARC Data Center.

Stratospheric processes

Gravity wave processes and their parameterisation

K. Hamilton reported on a 2-week school held in Trieste, Italy on the "Physics of the Equatorial Atmosphere" (see report in this issue).

R. Vincent announced that results from the H.R. radiosonde initiative extended to the S.H. was presented at IAMAS and that the work should be largely completed in 2002 with the submission of several papers to a special JGR section and the organization of a workshop. K. Hamilton described DAWEX (Darwin Area Wave Experiment). An Intense Observational Period (IOP) was ongoing while the SSG was in session, corresponding to the period when Hector (regular occurrence of intense convection) was occurring almost daily and often reached the stratosphere. He described the instruments involved, showed preliminary results and indicated how it will help in preparing for the larger scale ETCE (Effects of Tropical Convection Experiment) campaign.

R. Vincent and K. Hamilton presented and discussed with the SSG the plans for the next 3 years. The analysis of DAWEX data will likely span 2002-2003. They expect most papers resulting from DAWEX (including an overview paper) to be submitted by the end of 2003. Then the gravity wave initiative should continue to be involved in planning for a much larger field experiment (ETCE) to study effects of Hector convection in 2005 or later.

Future efforts will focus on reviewing existing data and encouraging appropriate new observational and modelling projects to characterise the spectrum of gravity wave momentum fluxes, including its geographical and seasonal dependence and its short-term intermittency. The aim will be to distil the available observational data and limited-area model results to provide as much guidance as possible for the formulation of source specifications and saturation mechanisms for parameterisation schemes. The gravity wave initiative will concentrate on this aspect, while leaving the main focus of gravity wave parameterisation to GRIPS.

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They proposed to initiate two specific activities. First will be a fairly large (about 70 participants) Chapman Conference in late 2003, which would initiate a general discussion of gravity wave parameterisation issues and which could be considered a follow up to the 1996 NATO meeting in Santa Fe. This could possibly be a joint activity with GRIPS, and second to hold a much smaller workshop in 2004 similar in scope and spirit to the 1998 La Jolla workshop on the QBO. This workshop would be focussed on producing a critical review/assessment of our knowledge of the gravity wave spectrum and the implications for practical parameterisation. The "deliverable" would be a refereed journal article.

Lower stratospheric/ upper tropospheric processes

This project concerns the transition region between the troposphere and stratosphere. The separation in time and space of chemical, radiative and dynamical processes in this region is not feasible as they are all strongly coupled. This region's key features are very low temperatures, high gradients and variability of water vapour, ozone and other species. Understanding the processes involving these species is of high priority for both understanding

climate and long-term changes in ozone. By an integrated approach this project looks at the processes that occur within and the interactions between the UT and the LS.

The main highlights in SPARC activities on this issue have been the publication of the review paper on "Atmospheric chemistry of small organic peroxy radicals" (Tyndall *et al.*, JGR, 106, D11, 12157-12182, 2001, see also report in Newsletter N° 15), the organisation of 2 workshops on Atmospheric Chemistry and the SPARC Tropopause Workshop.

A.R. Ravishankara, mentioned that the Tyndall *et al.* paper is being used for the NASA/JPL evaluation and will be used for the IUPAC data evaluations, thanks to the help of the SPARC Data Centre. Workshops were also held in the framework of joint SPARC-IGAC activities on chemical processes in the UT/LS, at the University of Heidelberg in March 2001 (see report in this issue) on Nitrogen oxides in the LS and UT, and at Breckenridge, Co, USA on laboratory studies of UT/LS processes, in July 2001 (the report will be published in Newsletter N°19).

T. Shepherd reported on the workshop, which took place in Bad Toelz, Germany, in April 17-21, 2001, and which was organised under his leadership together with P. Haynes, (see report in SPARC Newsletter N° 17). As already mentioned in this early report, the workshop gave the opportunity for intense debates on this very important region of interface between the stratosphere and the troposphere and help formulating key questions on the issue of chemistry/climate interactions. A review paper is being prepared which should be useful in defining further research that should feed into the SPARC/IGAC chemistry-climate activity.

Other scientific issues

Dynamical coupling of the stratosphere and troposphere

An update on recent developments on this issue was given by M. Baldwin and A. O'Neill. The apparent coupling between the stratosphere and the troposphere, as indicated by correlations in time series of the so-called Arctic Oscillation (AO), remains a subject of considerable interest and debate. By selecting time series of AO amplitudes after strong stratospheric warmings, M. Baldwin has shown evidence of

apparent downward propagation of anomalies from the stratosphere to the troposphere, implying that knowledge of the state of the AO in the stratosphere could increase predictive skill for the troposphere. A. O'Neill showed results (derived by A. Gregory) that give a measure of support to this suggestions. The potential improvement in predictive skill was shown, however, to be small though statistically significant. It seems likely that very strong events dominate the statistical results, and ensemble modelling experiments are needed to understand causal connections and mechanisms.

The clarification of stratosphere-troposphere links exhibited by variability in the AO signal will be a major focus of research for SPARC, in collaboration with other projects of WCRP.

A presentation of SOWER (Soundings of Ozone and Water in the Equatorial/Pacific Mission) results was given by M. Shiotani. The project has been strongly encouraged by SPARC since its beginning; it took place in San Cristobal-Galapagos, from 1998 to 2000. Besides presenting very interesting results, M. Shiotani raised the question of the possibility for upper air observations of San Cristobal as one of the GUAN stations being shut down in the near future and the SPARC SSG approved a letter to be written urging that station operations be continued at San Cristobal.

Chemistry-Climate interaction

Much of the IGAC and SPARC research agendas have their ultimate application in understanding chemistry-climate interactions. For instance, a central problem in stratospheric-climate interactions is to predict how polar stratospheric ozone will evolve in the future, taking account of increasing greenhouse gas concentrations together with decreasing "effective chlorine" resulting from the actions of the Montreal Protocol and its subsequent amendments. Substantial differences exist in present model predictions probably resulting from differing planetary wave transports in the future stratosphere.

In the troposphere, a central problem is to model the future greenhouse warming from troposphere ozone, methane, etc. In reality, of course, predicting how the future vertical structure of atmospheric ozone from the troposphere through the stratosphere is desired. Another problem is to predict how the changing atmosphere

might lead to changing upper troposphere-stratosphere water vapour concentrations. This will feed back on troposphere-stratosphere chemistry, which in turn will affect tropospheric climate. Cloud microphysics in the UT/LS is also a key issue, as it plays an important role in radiative forcing of the troposphere.

To more effectively attack these chemistry-climate problems will require the active participation of both the IGAC and SPARC communities. As a first step, it is proposed that SPARC and IGAC jointly convene a chemistry-climate workshop with the goal of developing a IGAC/SPARC chemistry-climate research agenda. The hope is that this research agenda can be advanced under the joint auspices of SPARC and IGAC.

Review of overall SPARC strategy and status of implementation

Understanding stratospheric climate change

The proposal of a new integrated SPARC initiative, with a view towards "Understanding Stratospheric Climate Change (1979-1998)", was discussed at length during the 2000 SSG meeting, under the leadership of V. Ramaswamy. The implementation was discussed further at the 2001 SSG.

The activity foreseen for SPARC in the future will aim to understand the observed stratospheric trends of temperature, ozone and water vapour, and solar effects, through modelling studies. These would be particularly aimed at elucidating UT/LS variability and its role in the overall climate system by building on the modelling work carried out in the stratospheric temperature trends study and GRIPS. New SPARC initiatives on stratospheric data assimilation are being undertaken. Within this framework, the main priority for SPARC is to continue to facilitate research on stratospheric processes and their role in climate by providing a forum or umbrella for international co-operation and encouraging inter-disciplinary exchanges. With the availability now of a sizeable body of information on the parameters of relevance describing stratospheric changes over the last two decades, it is appropriate to ask the following scientific questions:

- Are the different observed variations providing a consistent picture of stratospheric climate variations, including

the possibility of a trend over the past two decades, upon which shorter time scale variations are superposed?

- Can model simulations, employing the known forcings that have acted upon the system over the past two decades, be used in conjunction with the observed data to reproduce the changes in the observed parameters, and thereby lead to identification of the causes of these changes?

- Do conditions and processes in the stratosphere have an effect on tropospheric climate down to the surface?

The principal motivation arises from the fact that, over the period from the late 1970s to 2000, a number of changes, some dramatic, have been witnessed in the stratospheric climate: changes in ozone that are not the same from one decade to the next; aerosols from two volcanic eruptions perturbing the chemical and radiative budgets; water vapour data that indicate changes of various sorts depending on the times considered; pronounced temperature changes but with varying trends over the low and the middle and high latitudes, punctuated by sharp transient warmings in the aftermath of the volcanic eruptions; solar irradiance changes conforming to the 11-year cycle variations; variations in stratospheric circulation; and climate changes in the Arctic and Antarctic stratospheres especially during the winter-spring seasons.

Additional motivation arises from the fact that the coupling of the stratospheric and tropospheric climatic states is gathering increasing interest. The possibility that the tropospheric modes of variability may be related to stratospheric ones, and vice versa, is of considerable significance for climate change in the overall Earth system.

Thus, a SPARC-centred focus on understanding stratospheric change could have a direct bearing on changes from the upper troposphere down to the surface. The answers to these questions would also constitute useful inputs to the IPCC and WMO assessments. SPARC is uniquely placed to tackle the questions.

Interactions with other programmes and activities

SPARC maintains strong links and/or interacts widely as appropriate and necessary with several other projects of WCRP (WGNE, WGCM) and IGBP (IGAC). It was however noted that more cooperation should be established with CLIVAR and ACSYS, mostly

on the Arctic Oscillation. Particularly noteworthy is the joint SPARC-IGAC activity on UT/LS chemical processes which has been the best example of cooperation up to now. The proposed joint action on Chemistry-Climate interaction, which is going to be discussed further at the IGAC planning meeting in January 2002, should lead to an enlarged overlap between the two projects.

Reference was also made to the collaboration with SCOSTEP on upper stratospheric temperature trends and on the issue of solar influence on climate. The joint SPARC/SCOSTEP working group on the first issue will start its activity at the K lungsborn meeting in May 2002.

J. Gille presented the plans for the next COSPAR Scientific Assembly to be held in Houston, Texas, USA in October 2002 and where several sessions will be closely related to SPARC (see announcement in this issue).

M.-L. Chanin informed the SSG that the project IGACO (Integrated Global Atmospheric Chemistry Observations) was finally accepted by IGOS (Integrated Global Observing Strategy). This will be the opportunity to create an intellectual framework for integrated research in tropospheric and stratospheric chemistry. A committee under the chairmanship of G. Brasseur has been put in place and met for the first time in November in Geneva, Switzerland. The write up of IGACO will be largely based, at least for the stratosphere, on the report written for the WMO/CEOS Report (WMO N  140) entitled "Strategy for integrating satellite and ground-based observations of ozone".

M. Kurylo, SPARC ex Officio member representing NDSC, was unable to attend but sent his deep regrets and a message describing a possible interaction between NDSC and SPARC. He mostly suggested the publication in Newsletter N  19 of the summary of the NDSC Symposium which was held in Arcachon in September 2001, as well as the publication of a report from a recent workshop on Spectroscopic Needs for Atmospheric Sensing held in San Diego in October 2001. This was accepted without hesitation and the establishment of a closer link between SPARC and NDSC was welcome by the SSG.

The SPARC Data Center

The SPARC Data Center, supported by NASA, operated by P. Udelhofen at the State University of New York at Stony Brook has been continuing

assembling key stratospheric data sets in a readily accessible form. Established in June 1999, the number of data sets is growing rapidly. Several data sets are now online. Reference data sets based on UARS measurements and model analyses are available. High-resolution temperature and wind data from radiosondes, which were purchased from NOAA, are currently available for 1998, but will be augmented in the near future. Solar forcing and historic ozone data have been acquired. Data from the GRIPS model intercomparisons are now available. The Water Vapour Assessment (WAVAS) archive includes H₂O data from ground-based, airborne and satellite instruments.

The new data sets added this year include rocket data and the small organic peroxy radicals data. (<http://www.sparc.sunysb.edu/>).

The SPARC Office

As well as its regular activities (compiling and editing SPARC Newsletters, updating the SPARC mailing list, maintaining contacts with the SPARC community of scientists, organising various SPARC meetings and maintaining the SPARC home page), the SPARC Office had a large production during the year 2001. The SPARC water vapour assessment report was published in February 2001. The Proceedings of the SPARC 2000 General Assembly was produced as a

CD-ROM in March 2001. A new SPARC Brochure and posters were prepared and edited for the Global Change Conference of Amsterdam in July 2001.

Next SSG meeting

S. Yoden invited the SSG to meet in Kyoto, Japan in November 18-21, 2002, the week following the international symposium on "Stratospheric variations and climate" which will be held in Fukuoka, in November 12-15, 2002. The first circular for this symposium can be found on the WEB page: <http://fx.geo.kyushu-u.ac.jp/IntSympo/1st.html>. The invitation for both the SSG and the symposium was welcomed unanimously.

Next SPARC General Assembly

Five invitations had been received to hold the next SPARC General Assembly in 2004. The invitations came from:

- Dr Gufran Beig, for a General Assembly to be held in Pune, India.
- Dr Brian Connor, for a General Assembly to be held in Queenstown, New Zealand.
- Pr Daren Lu, for a General Assembly to be held in Beijing, China.
- Pr Norman McFarlane, for a General Assembly to be held in Victoria (BC), Canada.
- Dr Eugene Rozanov, for a General Assembly to be held in Davos, Switzerland.

The SSG was very impressed by such a manifestation of interest and there was considerable debate before making the final decision. The choice was motivated by the strong interest for SPARC to work more closely with the climate modelling community at a time when the stratospheric models are being implemented in the Climate GCM's. That led to the enthusiastic acceptance of the invitation from the Canadian Centre for Climate Modelling and Analysis (CCCma) of the University of Victoria, and to hold the General Assembly in Victoria, British Columbia, Canada. The date of the General Assembly is now fixed in July 19-23, 2004.

Announcement

A New Journal: Atmospheric Chemistry and Physics

Atmospheric Chemistry and Physics (ACP) is dedicated to the publication and discussion of studies of the atmosphere and the underlying chemical and physical processes. It is published by the European Geophysical Society, with an international Editorial and Advisory Board.

An innovative two-stage publication process ensures rapid peer-reviewed publication of papers, initially on the Atmospheric Chemistry and Physics Discussions (ACPD) web site. To foster scientific discussion, reviewer comments, short comments by other interested parties, and author replies appear alongside the original paper. Fully revised papers are published in ACP in both on-line and print versions.

On-line registration and subscription to ACP is free. It is financed on a non-profit basis by modest page charges. Colour graphics, supplementary material, and animations can be included at no extra cost.

ACP has been launched on 3 September 2001. To submit, view, or comment on papers, and for further information, please visit the web site: <http://www.atmos-chem-phys.org>.

An Update of Stratospheric Temperature Trends

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Introduction

The SPARC stratospheric temperature trends analyses have now been extended through to the year 2000. This represents an update of the work reported in WMO (1999) and Ramaswamy *et al.* (2001). While a focus in the earlier work was on the trends over the period 1979-1994, we present in this paper updated trends for the period 1979-2000, and compare the changes in the decadal-scale trend estimates for the two time periods.

In contrast to the data available from various platforms for evaluating the 1979-1994 trends, SPARC updates beyond 1994 have been made possible for the Angell, Berlin and Russia sonde data sets, the (Nash) SSU and MSU satellite data sets, and the CPC analyses (see Ramaswamy *et al.*, 2001, for a description of the various data sets). The SSU time series extends only through 1998. Also, the SSU data extends only up to ~70 degrees latitude in contrast to MSU which extends to 90 degrees. We summarise the trend results from the latest analyses performed as part of SPARC Stratospheric Temperature Trends Assessment (STTA), following closely the style of discussions reported in the earlier work. Updated trends estimates are computed using the maximum likelihood estimation methodology, in the same manner employed earlier to determine the 1979-1994 trends. The methodology consists of fitting the time series of monthly-mean values with a constant and six variables (annual and semi-annual sine, annual and semi-annual cosine, solar cycle and linear trend). The derived trend and standard error are the products of the computation. We describe here the estimates of the mean trends and the 2-sigma uncertainties resulting from the STTA analyses. As pointed out in the earlier paper, estimates of statistical uncertainties could be more sensitive to details of the trends determination method employed than the mean trend results themselves.

Results

Figure 1 (p. I) (left panel) illustrates the 1979-2000 trends in the zonal, annual-mean lower stratosphere temperatures,

as obtained from the MSU and SSU 15X satellite data sets. The right panel shows the 1979-1994 estimates from the same two satellite data sets (see also Plate 1 in Ramaswamy *et al.*, 2001). Also plotted are the 2-sigma level uncertainties in the trends for each time period. Considering each satellite data separately, the trend estimates in any latitude belt do not change appreciably, to within statistical uncertainty, in going to the longer time period. For both satellite data, differences in the mean trend values between the two time periods tend to be larger at the higher latitudes; for MSU, such differences are considerably smaller than for SSU. The SSU mean trend estimates are more negative for the longer period, especially in the mid-to-high latitudes in both hemispheres. For MSU, there is a slightly lesser cooling trend in the high northern altitudes for the longer time period, with the difference not being significant. For the 1979-2000 period, the SSU mean values are considerably colder than the MSU in the high latitudes of both hemispheres. Differences between MSU and SSU may be partially attributable to the different height regimes from where the signals are received for the two sensors (see Ramaswamy *et al.*, 2001). The peak in the SSU 15X signal emanates from a higher altitude than in the case of MSU. The slightly shorter record of the MSU could also be a contributing factor. The large variance seen at the equator, especially for the SSU, is likely due to the fact that QBO variance, an intrinsic feature of the climate system, has not been explicitly factored in the trends determination. Overall, the uncertainty estimates for the MSU and SSU are such that a consistency can be said to exist in their respective trends, over each latitude belt, in both hemispheres, and for each time period considered, irrespective of whether the trend at any latitude is statistically significant or not. Considering that the MSU is a microwave-sensing instrument and SSU an infrared one, it is noteworthy that both satellites indicate approximately similar trends, within the realm of statistical uncer-

tainty. Of particular note is the fact that, over both periods, the two satellite data sets indicate a statistically significant trend to be present in the mid-latitude (~30-60°N) Northern Hemisphere (NH) (see also Ramaswamy *et al.*, 2001 for the 1979-1994 trends discussions). It is somewhat encouraging that this continues to be a robust feature for the longer time period considered. It is further interesting that, in contrast to the shorter period, the 1979-2000 trends more clearly indicate the presence of statistically significant cooling trend in the Southern Hemisphere (SH) poleward of 20 degrees. For both data, MSU in particular, the uncertainty estimates are less for the longer time period.

Figure 2 (p. I) illustrates the zonal, annual-mean trends, and the 2-sigma uncertainty for the 50 hPa level, as obtained from the non-satellite data sets. As with the satellite data (Figure 1), in an overall sense, almost all latitude belts in the zonally averaged sense have a mean trend that is generally one of cooling. Considering the entire 1979-2000 period, the zonal pattern of the mean trend values in Figure 2 is seen to be generally consistent with the pattern for the satellite data sets in Figure 1, with a relatively larger cooling in the high southern and northern latitudes, a statistically significant cooling in the mid-latitudes, and a cooling trend that is accompanied by a larger uncertainty in the equatorial and high latitudes. In the SH, from the CPC analyses, the longer period exhibits an increased cooling trend than over the earlier period; however, the 2-sigma estimates remain large enough to suggest that it is difficult to discern a statistically significant change between the two time periods, as in the case of the satellite trends. At the equator, the sonde data sets do not indicate statistically significant results for either time period although there is hint of a lesser ambiguity for the longer time period. The large variation at the equator is, especially for Berlin over the 1979-1994 period, as in the case of the satellite data sets, likely due to not accounting for QBO in the trend estimation analyses. Between ~15-30°N,

the data sets suggest more of a tendency for a statistically significant cooling trend than over the shorter period. The Angell mean trends are slightly colder than Berlin in the low latitudes for the 1979-1994 period, but the agreement between them is considerably better for the longer period. In the northern mid-latitude (30-60°N), all the data sets tend to yield a statistically significant cooling over both time periods. For each data set, both the mean trend and 2-sigma estimates computed for the northern mid-latitudes using the additional years are remarkably consistent with the earlier determination for the 1979-1994 period. Coherency amongst the data sets regarding this feature is excellent for the longer period, too, just as was noted for the 1979-1994 period (Ramaswamy *et al.*, 2001). Added to the similar information content for these latitudes emerging from the satellite data (Figure 1), there is a robustness to the idea of an unambiguous cooling of the mid-latitude northern stratosphere over the past two decades. It is to be noted that the CPC data suggests a statistically significant cooling over almost all the SH belts for the longer period, consistent with the satellite data (Figure 1). Poleward of 60°N, the statistical significance is somewhat degraded in all the data sets for both time periods, primarily due to the increase in the dynamical variability. For each data set, there is a suggestion of lesser uncertainty for the longer time period in the high latitudes (see also Figure 1). There is an extremely good consistency between the Berlin, Russia and CPC data sets at 70 and 80°N for both periods. Near the pole, all data sets yield a large uncertainty yet they are mutually consistent in indicating a larger cooling there than at 60°N. The trend values in the high northern latitudes are not distinguishable in a statistical sense between the two time periods, consistent with the SSU and MSU results (Figure 1). From the sonde data, a statistically significant cooling becomes more discernible for almost all latitude belts (an important exception is the high northern latitudes) over the longer period, consistent with the feature seen in Figure 1 for the satellite data. The good agreement between the data sets strongly confirms the notion of an unambiguous cooling of the global lower stratosphere over the last two decades, with the area of statistically significant cooling being more extensive than just the northern mid-latitude regions as was noted for the 1979-1994 period (WMO, 1999).

Figure 3 (p. I) illustrates the annual-mean vertical profile of the trends in the NH averaged stratosphere (100 to 1 hPa) over the 1979-1994 period from sonde (Berlin and UKRAOB) and satellite (MSU and Nash SSU) data sets. The satellite values (MSU and SSU) have been plotted at the altitude of the peak emission altitude whereas, in actuality, the estimates represent the trend over a broad altitude layer (see Ramaswamy *et al.*, 2001). In the lower stratosphere (30-100 hPa), the various data sets exhibit consistency in terms of both the mean trends and the 2-sigma values. Thus, throughout the lower stratosphere, not only can it be asserted that a statistically significant cooling has occurred, but that this is consistent for the different instrumental data. The mean cooling trend estimates in the 100-50 hPa and 30 hPa region are ~ 0.30 - 0.75 K/decade and 0.9 K/decade, respectively. The presence of a statistically significant trend, however, should be distinguished from the fact that there is a non-negligible spread in the estimate from any data set at any of the lower stratospheric altitudes, with the uncertainty being at least ~ 0.5 K. This hints at a limit to the accuracy with which the stratospheric decadal-scale changes can be pinned down currently using the 95% level confidence estimates. Above 30 hPa, the only values that are available globally are the Nash SSU trends. The vertical trend profile confirms the general picture expressed earlier for northern mid-latitude (45°N) (Ramaswamy *et al.*, 2001) meaning that there is a decrease in the cooling trend occurring in the middle stratosphere, with a minimum (~ 0.25 K/decade) occurring at ~ 5 hPa. Above this altitude, there is an increasing cooling trend with altitude through to the stratopause (~ 1.2 K/decade at 2 hPa). The NH annually-averaged cooling is statistically significant from 100 to 1 hPa. It is unfortunate that there is a critical lack of other data sets that can provide corroborative information on the global trends in the middle and upper stratosphere, a limitation that constrains slightly the robustness of the currently-derived trend values in the upper stratosphere, especially when contrasted with the situation for the lower stratosphere.

Discussions

The NH annual-mean lower stratospheric (30-100 hPa) temperature trends (1979-1994) are statistically significant and exhibit a remarkable degree of consistency in terms of the trends and

the 2-sigma estimates amongst the different radiosonde and satellite data sets. In the middle and upper stratosphere, there is only one data set that is available to estimate hemispheric trends. Nonetheless, the NH stratosphere, in an annually averaged sense, displays a statistically significant cooling from the lower through to the upper stratosphere. The shape of the trend in the vertical profile of temperature decrease is an important and distinct aspect of climate change over the past two decades. It ranks as a major, statistically significant change discerned in the climate variables for the 1979-2000 time period. Notwithstanding this determination of an unambiguous cooling, it is important to bear in mind the uncertainty in the trend estimates when pursuing the attribution issue. For the zonally-averaged lower stratosphere, the annual-mean radiosonde trends over the 1979-2000 period indicate a statistically significant cooling over almost all the mid-to-high latitude in the two hemispheres, with the satellite data, too, hinting at this feature over all areas except the high northern latitudes. This inference is an important update to the conclusion in WMO (1999) and Ramaswamy *et al.* (2001) with regards to the 1979-1994 trends, when the statistical significance of the cooling in the 50-100 hPa region was confined to only the northern mid-latitudes. There are suggestions that a longer period of evaluation yields the tendency for a statistically significant cooling even at low latitudes. A realistic assessment of southern latitudes is somewhat hampered by the lack of availability of an adequate number of data sets, in contrast to the case for the NH.

With the observation-derived trends on the hemispheric and zonal, annual-mean time scales on a firm footing, there is a challenge to explain the causes of the observed temperature variations and changes on these space-and-time scales. In particular, it is important to address the question of the relative roles of well-mixed greenhouse gases, ozone, water vapour and aerosol variations/changes upon the evolution of the stratospheric temperatures, in part to assess the overall part played by externally applied radiative forcings in affecting stratospheric climate. In this regard, careful formulation and analyses of model simulations are needed, which, when compared with the observations, can provide a resolution of the attribution issues, and quantify the parts played by natural and anthropogenic factors. The models that are being deployed to

address these challenges include general circulation models (GCMs) in which the trace species changes are prescribed, to coupled chemistry-climate models where the evolution of species is prognosed in some manner. While the roles of trace gases have been studied extensively and have likely been identified successfully as being the principal cause of the global-mean decadal-scale trends in the ~50-100 hPa region (WMO, 1995 and 1999), the precise roles of all trace species in explaining the vertical profile of the changes in the entire stratosphere

has yet to be made definitive. Eventually, the modelling exercises will need to advance further and explain the cause/s of temperature evolution on spatial scales that are finer than zonal, and on time scales that are finer than annual.

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Assessment of Stratospheric Aerosol Properties (ASAP)

Report on the Assessment Kick-Off Workshop, Paris, France, 4-6 November 2001

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Aerosol is one of the most variable components of stratospheric air. As the result of episodic volcanic eruptions, the mass of aerosol in the stratosphere has varied by about 2 orders of magnitude over the last 25 years. Aerosol in the upper troposphere/lower stratosphere (UT/LS) can have a significant impact on climate through radiative effects and on stratospheric chemistry, particularly impacting ozone. The magnitude of these effects is also highly variable and, as a result, can complicate isolating human-derived changes in the stratosphere. Accounting for aerosol effects properly is a key component of modelling climate/chemistry effects properly.

By mass stratospheric aerosol consists of almost exclusively aqueous sulphuric acid droplets. However, its precise characterisation is nonetheless complex. The details of composition and size distribution strongly influence important aerosol properties like

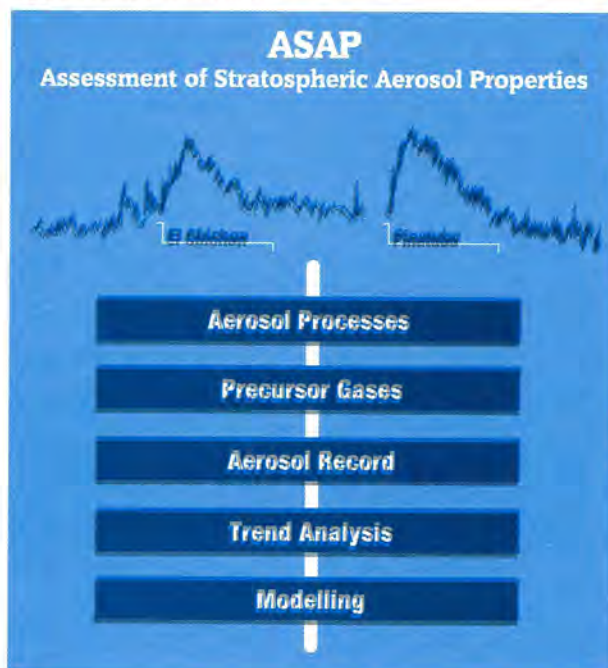
surface area density and the spectral dependence of aerosol scattering and absorption. While most gas species measurements, such as those of ozone, are essentially measurements of the species molecular density and hence

by itself useful, aerosol instruments employ a variety of measurement strategies, each determining only some aspects of the underlying aerosol distribution. As a result, the conversion of measurements to practical parameters generally requires model-based transformations that may themselves be subject to substantial uncertainty.

In recognition of the importance of stratospheric aerosol, SPARC has initiated a project to evaluate the scientific understanding of UT/LS aerosol and aerosol measurements. The assessment was initiated with a workshop in Paris, France on 4-6 November 2001, supported by the SPARC Office. Thirty aerosol scientists attended the workshop and identified key scientific issues that should be addressed in the assessment. These include:

- How representative are satellite-based aerosol records and derived climatologies?
- How have key aerosol properties (e.g., surface area

Figure 1. ASAP Structure



density) and precursor gas loadings varied with time?

- What is the non-volcanic background for stratospheric aerosol? Can a trend in the background level be inferred?

- How well does the aerosol record/climatology reflect our understanding of aerosol processes, in particular in regions of or at times of enhanced aerosol nucleation, coagulation, growth and evaporation?

- How well can models using gaseous precursors to produce observed aerosol properties under non-volcanic and volcanically perturbed conditions?

The assessment will attempt to address these questions in five working groups: processes, aerosol precursors, aerosol record and climatology, trends, and modelling. In parallel with the construction of the aerosol record, that we intend to become the most comprehensive one compiled so far, the assessment of precursor gases, aerosol trend detection, and aerosol modelling will proceed with regular feedback between groups to ensure consistency. These activities are also hoped to merge into an aerosol climatology reflecting stratospheric aerosol properties during volcanically quiescent periods.

The aerosol processes group will review our knowledge of the aerosol lifecycle including their formation in and removal from the stratosphere, thereby providing context to all sections of the assessment. Specific issues to be addressed include the nucleation of aerosols at the tropical tropopause, and in the free stratosphere following volcanic events. Processes that control clouds in the vicinity of the tropopause and polar stratospheric clouds will also be reviewed. The aerosol precursors group will quantify what is known about the non-volcanic gaseous precursors to stratospheric aerosol formation and produce a database for further experimentation. This includes natural and human-derived source species such as OCS, tropospheric SO₂, and DMS. This group will also address whether there are discernible trends in the gas concentrations, possibly of anthropogenic origin.

The primary products of the aerosol record will be satellite-based and as such, the primary parameters will be aerosol extinction at a variety of wavelengths. Key secondary products (e.g., aerosol surface area density and effec-

tive radius) will be derived and supplemented with additional parameters (e.g., conversion factors between backscatter and extinction at a few wavelengths). We expect that the long records from SAGE II, HALOE, and other space-based instruments will play a prominent role in building a comprehensive data record, while balloon-borne and lidar records will further serve to validate the remotely estimated values. The construction of the climatology for volcanically quiescent periods will depend crucially on *in situ* observations like the University of Wyoming optical particle counter (OPC) and on long-term lidar measurements like the Garmisch record. Since there is evidence for significant systematic differences between some satellite and *in situ* measurements in derived quantities, like surface area density, we will address the potential for bias and limitations in general.

The record will be compiled as a function of latitude, altitude and time, and possibly also as a function of equivalent latitude, potential temperature and time. The altitude range of interest extends from 2-3 km below the tropopause to the stratosphere. Hence, it will include the scientifically interesting and for satellite instruments easily accessible region of the uppermost troposphere. This spans a region in which ice clouds frequently occur. We intend to quantify the frequency of occurrence of cirrus events as a function of time and location but otherwise exclude them from aerosol data products. The final products will be electronically available through the SPARC Data Center.

The evaluation of trends will concentrate on the volcanically quiescent periods, though much can be learned about natural aerosol variability in volcanically perturbed periods as well. Underlying data include aerosol mixing ratios from the OPC, backscatter ratios from the lidars, and aerosol extinction for HALOE and SAGE II. The trends group will determine what periods can be identified as being at non-volcanic levels, if any. The cleanest periods, for which substantial data are available, are the late 1970's, the late 1980's and the late 1990's through the current time. These periods will be compared using a rigorous statistical approach, and the significance of any observed differences will be evaluated.

The modelling group will bring together the measured aerosol source gas budgets, the observed volcanic and background aerosol levels, the derived trends in the latter periods, and the modelling of transport, oxidation, nucleation and condensation processes. While this group is not intended to perform a comprehensive comparison between global aerosol models, the agreement between the observed aerosol and that produced by various models is expected to give significant insight in the how close we are to understanding the processes that control aerosol in the stratosphere.

Acknowledgements. We wish to thank the many colleagues who have accepted to serve as authors in this assessment and whose enthusiasm for the topic was evident throughout the Paris workshop. We would further like to thank CNES for hosting the meeting, and most prominently the SPARC Office, whose nonpareil support in organising this workshop was a key element in its success.



Announcement

The 6th European Symposium on Stratospheric Ozone

The Sixth European Symposium on Stratospheric Ozone will be held at Chalmers University of Technology, Göteborg, Sweden from 2nd to 6th September 2002. As in the previous workshops at Schliersee and St-Jean-de-Luz, this symposium will be a major opportunity for the presentation and discussion of results from the European Union research programme on stratospheric ozone. Any questions to Rebecca Penkett at general@ozone-sec.ch.cam.ac.uk or see:

<http://www.ozone-sec.ch.cam.ac.uk/>

Report on the Nitrogen Oxides in the Lower Stratosphere and Upper Troposphere Workshop

University of Heidelberg,
Heidelberg, Germany, 19-22 March 2001

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Introduction

Reactive nitrogen oxides play key roles in the photochemistry of the upper troposphere and lower stratosphere (UT/LS). Significant progress has occurred in the last decades in both the measurement and modelling of reactive species and their distribution in the UT/LS. The sampling of the UT/LS has increased with the addition of new instruments on space borne and airborne platforms. Models have advanced to three-dimensional chemical-transport models and more precise and accurate trajectory models. Significant advances have also been made in quantifying rate coefficients of gas-phase, heterogeneous, and photochemical processes in the laboratory. This workshop was convened to address the recent progress in measurements, and modelling and to highlight areas of research in which significant uncertainties remain.

The Workshop was convened by G. Amanatidis (European Commission, Brussels, M. Chipperfield (University of Leeds, UK), R. A. Cox (University of Cambridge, UK), D. W. Fahey (NOAA Aeronomy Lab, Boulder, USA), N. Harris (EORCU, University of Cambridge, UK), Y. Kondo (University of Tokyo, Japan), M. J. Kurylo (NIST/NASA, Washington, USA), M. Pirre (CNRS Orléans, France), A. R. Ravishankara (NOAA Aeronomy Lab, Boulder, USA), through the initiative of Klaus Pfeilsticker of the University of Heidelberg who was also the local host. Approximately 65 people attended the three-day meeting on the University of Heidelberg campus. The meeting was organised with a core group of presentations followed by extended discussion sessions. Separate breakout sessions were held for modelling, observations, and laboratory measurements, and the meeting concluded with a rapporteur's report and summary for each of these topics.

Highlights of the workshop

In the following sections, the discussions and presentations of modelling and observations of reactive nitrogen in the

UT/LS are summarised. The discussions and presentations identified areas of high confidence, areas of limited confidence, and knowledge gaps/needs as shown in Table 1 (p. 12).

Modelling reactive nitrogen in the lower stratosphere

The principal component species of NO_y in the stratosphere are HNO_3 , NO_x , N_2O_5 , ClONO_2 , BrONO_2 , and HO_2NO_2 . NO_x is here defined as the sum of NO and NO_2 . Figure 1 (p. II) shows the photochemical interrelationship of these species. The most significant source of stratospheric NO_y is N_2O in reaction with $\text{O}(1\text{D})$ to produce NO and photolysis of NO is the primary *in situ* loss process of NO_y . Contributions from galactic cosmic rays, solar proton events and solar electrons are relatively small (<20%) on average. In the lower-most stratosphere, lightning and aircraft NO_x may contribute to the NO_y budget. NO_y is produced in the mid- and upper stratosphere and transported poleward and downward into the lower stratosphere (LS). NO_y is chemically conserved in the LS. The main sink of NO_y in the LS is cross-tropopause transport. Sedimentation of polar stratospheric cloud particles in the polar vortex can effect redistribution of NO_y within the LS and transport NO_y to the troposphere.

Modelling NO_y in the LS involves processes on very different time scales. The distribution of N_2O and NO_y depends largely on global transport, while, for example, the partitioning of NO_x species has a much shorter time scale and can in many cases be considered to be in photochemical steady state. The mean age of air in the high-latitude stratosphere is several years or more. The partitioning between NO_x and HNO_3 takes place on an intermediate time scale of about a few days. Modelling the distribution of NO_y thus requires 2D or 3D photochemical-transport models, while the partitioning within the NO_y family can often be

calculated by photochemical box models. Box models constrained by observations provide probably the best way to test our understanding of the NO_y chemistry (e.g. Gao *et al.*, 1999; Cohen *et al.*, 2000). In many cases, however, not all relevant parameters needed to constrain the model calculations are measured sufficiently. In these cases, global 3D photochemical-transport models constrained by meteorological analysis provide the best tool available to interpret these measurements.

Many aspects of the stratospheric NO_y chemistry are well understood. In particular, the principal species which constitute NO_y in the stratosphere are known (Figure 1). *In situ* measurements from the ER-2 aircraft in comparison with the Mk IV remote measurements have shown that the sum of the individual NO_y compounds is in good agreement with total NO_y (e.g. Sen *et al.*, 1998). It is unlikely that there are any significant unidentified NO_y components in the stratosphere. However, many models show relatively large discrepancies for the distribution of NO_y compared with observations, even when N_2O is constrained. In addition there are relatively large discrepancies for NO_y and N_2O between different models. These discrepancies are likely due to the transport features of the model and possibly the NO loss processes rather than the production of NO_y from N_2O decomposition.

The partitioning on short time scales that controls the NO/NO_2 ratio, for example, seems to be well reproduced by constrained models (e.g. Del Negro *et al.*, 1999). However, the agreement with observations may be less good for globally unconstrained models, as the NO/NO_2 ratio depends critically on ozone and the NO_2 photolysis rate. One issue here is the treatment of the albedo in global models. Sen *et al.* (1998) show good agreement between the observed N_2O_5 diurnal cycle and constrained model calculations.

Lower Stratospheric Modelling	Upper Tropospheric Modelling	Stratospheric and Tropospheric Observations
High Confidence		
• NO_y composition • Partitioning of NO₂/NO, ClONO₂ • Source of NO_y	• Principal NO_y species and their abundance • HO_x and NO_x partitioning • Aircraft NO_x emissions • Large-scale transport	• Stratospheric distribution of NO, NO₂, and NO_y • Correlations of NO_y with N₂O in the LS • NO₂/NO and NO_x/HNO₃ ratio over a wide range of conditions in the LS
Limited Confidence		
• Partitioning of NO₂/HNO₃ • NO_y transport with specific emphasis on cross-stratopause and cross-tropopause transport • Net NO_y yield from N₂O in model intercomparisons • Aspects of polar stratospheric cloud formation and growth	• Minor species abundance • NO_x lightning source • Small-scale transport and convection • Photolysis rates in the presence of cloudiness	• Distribution of non-NO_x species in the LS • Cross-tropopause transport and its effect on composition • Transport of NO_y in the stratosphere • Aerosol surface characterisation in the UT • Distribution of minor NO_y species
Knowledge Gaps/Needs		
• Distributions of HO₂NO₂, BrONO₂, N₂O₅ • NO_y transport in the tropopause region • Nucleation of polar stratospheric cloud particles	• NO_x/HNO₃ ratio • 2000 aircraft emission data base • Kinetic data for homogeneous and heterogeneous processes • Simulation of heterogeneous processes in models • Washout processes and rates • Long-lived tracer data to constrain transport models • Formal model/data intercomparison	• Trends in NO_y in the LS; cause of NO₂ trends in the LS • Correlations of short-lived tracers such as acetylene and CN, HO_x radicals (OH, HO₂ and RO₂) with their precursors such as acetone, HOOH and ROOH in the UT • Aerosol parameters necessary to describe HNO₃ (and perhaps other NO_y species) uptake efficiency on cirrus particles • Observations of global and seasonal distributions of NO_x, HNO₃ and PAN in the UT • Sources (especially lightning) and sinks of NO_y in the UT • NO_y partitioning • NO_y relationship to age of air in the LS
**Areas noted under Limited Confidence and Knowledge Gaps/Needs for Stratospheric and Tropospheric Modelling represent observational needs which are not repeated in the Observation entries.		

Table 1. Knowledge summary**

Similarly, Stimpfle *et al.* (1999) report reasonable agreement between observed and modelled ClONO₂.

The partitioning between NO_x and HNO₃ in the LS depends critically on the aerosol surface area. The partitioning between NO_x and HNO₃ is fairly well simulated in models which are constrained by observed aerosol surface area. Recent changes in reaction rate recommendations (e.g. Brown *et al.*, 1999a,b) have improved the agreement between observed and modelled NO_x/NO_y. Discrepancies are on the order of 10-20% at ER-2 altitudes (15-20 km) (Gao *et al.*,

1999). However, comparisons between observed NO₂ from SAOZ balloon-borne measurements and three-dimensional models show that at other altitudes the agreement is less good, with differences of the order of 50%. Jucks *et al.* (1999) have shown using far infra-red observations that constrained models underestimate NO_x and overestimate HNO₃, with the discrepancy increasing with altitude. One way to resolve this discrepancy would be to increase the HNO₃ photolysis near 200 nm. It remains an open issue how well models reproduce the UV flux at short wavelength in the stratosphere. Many models include paramete-

risations of the atmospheric transmission in the Schumann-Runge bands, which have not been optimised for HNO₃ photolysis calculation.

A critical parameter for modelling the partitioning between NO_x and HNO₃ is the concentration of OH. Both loss and production of HNO₃ depend on OH. Previous studies have shown good agreement between observed and modelled OH, in particular for the ratio of OH/HO₂. However, the most recent JPL 2000 reaction rate recommendations lead to large differences between modelled and observed OH/HO₂ ratio, with

observed OH about 50% higher than modelled (Lanzendorf *et al.*, 2001). Current models significantly overestimate the ratio of HO_2NO_2 to NO_x for high-latitude spring, compared to observations. This may represent a gap in our current knowledge. A speculative long-wavelength photolytic pathway for HO_2NO_2 has been suggested to resolve this discrepancy (Wennberg *et al.*, 1999).

Bromine nitrate (BrONO_2) is an important bromine reservoir and thus a critical component for modelling stratospheric bromine chemistry. The heterogeneous hydrolysis of BrONO_2 affects the partitioning of stratospheric NO_x and HO_x (e.g., Erle *et al.*, 1998). Unfortunately, BrONO_2 has never been measured in the stratosphere. The observed rapid increase of HO_x after sunrise is basically consistent with model calculations of the hydrolysis of BrONO_2 and subsequent HOBr photolysis and may thus provide some indirect evidence for BrONO_2 .

PAN is generally not included in stratospheric models. Its contribution to NO_y is believed to be small in the stratosphere but more significant in the tropopause region.

There was relatively little discussion at the workshop on formation of polar stratospheric clouds (PSC) and denitrification. Although models are able to predict some aspects of PSCs reasonably well and there has even been some success in modelling observed denitrification, many aspects of PSC formation are still uncertain. Very little is known about the precise PSC nucleation mechanism. This limits our confidence in modelling denitrification and the ability to predict the future evolution of Arctic ozone depletion in the presence of long-term trends of temperature and water.

Modelling reactive nitrogen in the upper troposphere (UT)

In the UT, NO_x and NO_y essentially are governed by many processes that are highly coupled (see Figure 2). As in the stratosphere, the concept of NO_y is a useful constraint and diagnostic. However, UT NO_y is more variable due to the variability and uncertainty in emissions, chemical processing, transport, and loss by wet and dry deposition. Therefore in the UT, it is often more important to measure and model individual species such as NO_x , HNO_3 , and PAN. The principal tropospheric NO_y species and their inter-conversion processes are shown in Figure 3 (p. II).

By contrast to the stratosphere, where chemical and transport time scales are often very different, both transport and photochemistry act on the distribution of NO_x , HNO_3 , PAN, and other NO_y compounds in the UT with similar time constants of weeks to months. Mixing ratios of UT NO_x and HNO_3 can be lower than those at the surface and in the LS. Cross-tropopause transport and convection are sources of NO_x and HNO_3 in the UT. Direct aircraft emissions of NO_x into the UT are also important, as is NO_x generated by lightning in the UT or at lower altitudes where it can be transported to the UT by convection. Net removal of NO_y compounds from the UT is primarily by conversion to HNO_3 followed by uptake into particles and sedimentation. One consequence of the coequal time scales for transport and photochemistry is that comparatively few species are routinely in 24-hour photostationary state within the UT. Diagnostics, such as the number of condensation nuclei or the abundance of other short-lived species, have been suggested as indicators of the residence time of air in the UT.

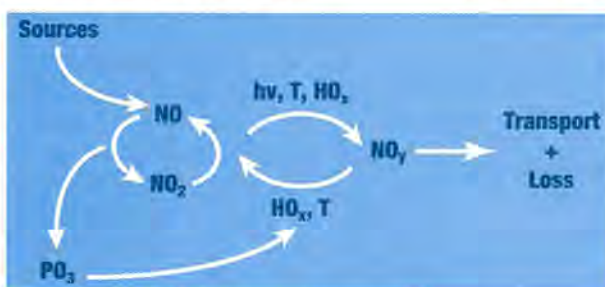


Figure 2. Schematic of upper tropospheric NO_y processes. [Courtesy of K. Law].

In the UT there are still many uncertainties and unknowns related to our understanding about NO_x and NO_y and associated chemistry. Two areas were identified where our knowledge is felt to be good. These include our understanding of fast gas-phase photochemistry involving HO_x and NO_x partitioning where the use of observationally constrained box models run to steady state has shown that we have a reasonably good understanding about some aspects of radical budgets in the UT. However, uncertainties still remain about the exact nature of HO_x sources and sinks and the rate of *in situ* O_3 production at high NO_x levels (see for example, Jaegle *et al.*, 1999). There is a lack in our ability to simulate NO_y partitioning, most notably the NO_x/HNO_3 ratio. Although several mechanisms have been proposed (e.g. reactions on soot) to address the discrepancy between observed and modelled ratios

(the latter are too low), no satisfactory explanation has been found to date. It was also felt that we have reasonably high confidence in the total amount of NO_x emitted by aircraft.

With respect to NO_x emissions, the distribution of lightning emissions in mid-latitudes is reasonably well known (Huntreiser *et al.*, 1998; Dye *et al.*, 2000). However, large uncertainties remain in describing the distribution of lightning in the tropics and the magnitude of NO_x emissions from this source, particularly in the tropics. Also, there are considerable difficulties in modelling this source due to the scales involved and the coupling to convection. The problem of including emissions, which are very heterogeneous in time and space, in large-scale global models which have grid boxes on the scales of 10-100s km is a problem which requires further attention, in general, particularly as the response of the chemical system is non-linear with respect to NO_x concentrations. Global inventories do exist for surface and aircraft emissions and the total amounts of NO_x emitted from some

sources (aircraft, combustion) are well known compared to those from other sources (biomass burning, soils). However, several data sets currently used by models require updating to more accurately represent emissions in

2000, for example. It was also felt that the stratospheric flux of NO_y into the troposphere is reasonably well known.

Modelling NO_x and NO_y in the UT (and elsewhere) relies on accurate kinetic and photolysis rate calculations. There is a clear need for more accurate data on reaction rates, cross sections and quantum yields under UT temperature and pressure conditions and for the mechanisms, processes, and rates of heterogeneous reactions. Related to the latter is the whole question of how to simulate heterogeneous processes in models. This topic is one that will require much more attention in the future. Also, whilst models are able to reproduce measured photolysis rates in clear sky conditions, there are still considerable problems reproducing data collected in cloudy and high aerosol conditions. This will require both new data and model improvements.

The ultimate fate of NO_y in the free troposphere involves washout of soluble species, such as HNO_3 , uptake on

aerosols/droplets/ice particles or transport downward to the surface and loss by dry deposition. These processes can also affect the partitioning between NO_x and NO_y , and are especially important if they regenerate active NO_x which is then available to produce O_3 in the troposphere. The whole question of loss of HNO_3 and other species by washout has received too little attention both in the modelling community and the measurement community largely due the complexity of the processes involved. Indeed, the treatment of clouds in models is still very crude and the treatment of wet deposition depends mainly on derived precipitation rates together with information on solubility. Measurements of nitrate in rainwater have been used to validate modelled wet deposition of HNO_3 but clearly more observational data and also laboratory data on rates and mechanisms are still needed.

The relative distributions of NO_x and NO_y in the UT depend on transport processes as well as emissions, chemical processes, and loss processes. Based on the comparison of global models with (rather limited) tracer data (e.g. CFC-11) and the improved reliability of current weather forecasting models, it can be concluded that our ability to model the large-scale transport in the troposphere is reasonably good. This also explains why back trajectory calculations have been used very successfully in recent years to study the origin of different air masses observed during measurement campaigns. However, many transport processes occur at smaller scales. These include frontal transport and stratosphere-troposphere exchange which are resolved by current models when run at high enough resolution although convection embedded in frontal systems and sharp gradients around the tropopause are often not resolved. Deep convection is parameterised in current chemistry-transport models. Whilst there have been improvements in recent years (e.g. see results from WCRP model intercomparison exercises, e.g. Jacob *et al.*, 1997), further improvements can be envisaged based on new parameterisations, increased resolution, and the use of nested models. These will clearly be necessary if we are going to be able to simulate fine-scale layers that are widely observed in the UT. At the present time it is unclear whether there is a need to model these features in order to simulate the global NO_x , NO_y , and O_3 budgets more accurately.

Further measurements of tracers that are very useful for validating transport processes in models would significantly improve our modelling capability (e.g. 222-Rn, 210-Pb, CO_2 , 7Be/10Be).

Finally, as a way forward, the idea of a formal model/data intercomparison was discussed. State of the art box models, including current chemical schemes could be compared to available NO_x and HO_x data from various airborne campaigns. Comparison with NO_y data and its individual components would require the use of global or regional models that take into account transport and scavenging processes. A main objective would be to test the sensitivity of current models to these processes over a wide range of conditions in the UT.

Observations of reactive nitrogen species in the UT/LS

Lower Stratosphere

There is high confidence that the distribution of NO_y within the LS is controlled almost entirely by transport from the middle and upper stratosphere and across the tropopause. The NO_y created at high altitudes within the stratosphere is then distributed into the LS by advection and diffusion on a time scale of years. In the overworld (defined as the stratosphere above the 380K potential temperature surface), there are extensive, high-resolution *in situ* observations of N_2O , the principal source of NO_y . Based on these observations, we have a good working knowledge of the N_2O distribution in space and time. In the overworld, observations have demonstrated that there is a tight correlation of NO_y with N_2O , except in air that has been influenced by recent PSC sedimentation, and that this correlation varies seasonally. For N_2O values less than about 50 ppbv, the $\text{NO}_y/\text{N}_2\text{O}$ correlation becomes non-linear and is associated with greater uncertainty.

By contrast to the distribution of NO_y which is set on a time scale of years, the partitioning of NO_y among the different species, HNO_3 , NO , NO_2 , N_2O_5 , ClONO_2 , etc., within the LS is established on a time scale of minutes to 30 days. Because transport of air in the stratosphere is approximately zonal, most of the photochemical relationships between NO_y species exist in a 24-hour diurnal photostationary state. The most important NO_y species and all the gas phase processes that interconvert them have been identified (see Figure 1).

Some observations have been analysed to show that we have sampled a broad range of one or two of the parameters controlling the NO/NO_2 ratio and the NO_x/HNO_3 ratio, but a narrow range in others. Where we have sampled a broad range in the parameters, we can provide a quantitative evaluation of the completeness and accuracy of model processes. For example, atmospheric observations at low temperatures imply that the uncertainty of the rate coefficient for $\text{NO} + \text{O}_3$ is 5-15% at 200K and by less than that over the range 200-220K. This uncertainty is significantly less than that currently derived from the extrapolation of laboratory measurements made at higher temperatures. Observations also indicate the rate ratio of the $\text{OH} + \text{NO}_2$ reaction to the sequence of processes that convert NO_2 to N_2O_5 and then to HNO_3 by heterogeneous reaction are accurately represented in models using current laboratory rate expressions.

While we have high confidence in our knowledge of the distribution of NO , NO_2 , NO_y , observations of NO_3 , N_2O_5 , HNO_4 , PAN, ClONO_2 , and HNO_3 are far fewer, and for the most part these observations have been made by remote sensing and lack the context of simultaneous, accurate free-radical measurements. Consequently the confidence in our knowledge of the distribution of these species and the chemistry affecting that distribution is limited. This is primarily important for HNO_3 because it can condense at very low temperatures. For the other species, the lack of observational tests means that our ability to demonstrate that we have identified all of the important processes and established that their rates are correctly modelled is not currently possible.

Observations of the composition of the middle-world (below 380K) are also not as extensive as observations of composition in the overworld. Our ability to describe the frequency and intensity of cross-tropopause transport and subsequent mixing on the composition in this region is limited.

An increasing trend has been observed in column NO_2 amounts over the last two decades. The most likely cause is related to NO_y aerosol chemistry or stratospheric transport. Concerning NO_y , we have little to no understanding or observational demonstration of whether or not trends in tropospheric N_2O are producing the expected NO_y trends in the stratosphere. Combined long-term records of NO_2 , HNO_3 , aerosol, N_2O , and the extent of denitrification are

needed. Observations should include sampling in the tropics, mid-latitudes, and the poles and at altitudes capable of resolving exchange between the UT and LS across the tropopause.

Although the new SOLVE observations (e.g. Fahey *et al.*) demonstrated the existence of a class of very large PSC particles with strong denitrification potential, information of their nucleation are still lacking. Additional studies confirming the generality of the new observations should be conducted.

There is considerable room for improvement in our understanding of the effects of stratospheric transport on the distribution of NO_y . Much of this is not specific to NO_y but associated with clear problems that have been identified in our ability to model the mean age and the age spectrum of lower stratospheric air. The mean age has been derived from the correlation of N_2O with quite extensive measurements of CO_2 and SF_6 , and the age spectrum from the CO_2 measurements. For NO_y specifically, observations and analyses of observations that would lead to an improved understanding of the seasonal cycle of NO_y and its relationship to both the mean age and the age spectrum in middle-world is needed.

The role of NO_y production by high-energy electrons associated with geomagnetic disturbances is unclear. Since the geomagnetic disturbances are closely related to the 11-year solar cycle, this production mechanism may cause a similar cycle in NO_y in the LS.

Comparison of HNO_3 chemistry in the lower and upper stratosphere points to possible errors in descriptions of the photon flux at short UV wavelengths ($< 310 \text{ nm}$). These could potentially be resolved by *in situ* actinometry to measure J_{O_2} , J_{NO} , $J_{\text{N}_2\text{O}}$ or J_{HNO_3} .

The use of chemical coordinates should be expanded to more precisely refine our understanding of fast photochemistry and to more clearly define which aspects of stratospheric NO_y science are dependent on transport and which on chemical parameters alone.

Aerosol surface area is a central control over the abundance of NO_y in the LS. Demonstration of a model capable of predicting from elementary principles the observed aerosol- N_2O correlations and/or the geophysical distribution of aerosol remains a challenge.

Balloon-borne, long-path UV-visible measurements have occasionally shown relatively high levels of NO_2 together with high levels of OClO in

the Arctic vortex. An analysis shows that these observations are not consistent with our current understanding of winter stratospheric chemistry. Further work will examine the role of vertical resolution and spatial inhomogeneity on the retrievals.

Upper Troposphere

The chemistry of the UT is more complex than the chemistry of the stratosphere because many more species are involved and the system is inherently much more heterogeneous with orders of magnitude variation routinely observed in NO_x , hydrogen radical precursors and H_2O . The sampling of the UT has increased significantly in the last decade due to an increased number of airborne sampling missions using instrumented aircraft in the US, Europe, and Japan; and due to the successful launch of new satellite instruments.

Airborne platforms are now frequently instrumented for a wide suite of reactive nitrogen measurements including NO_y , NO , NO_2 , HNO_3 , and PAN and occasionally for other organic nitrates. The sampling campaigns of these aircraft have made measurements over a wide range latitudes, longitudes, and seasons. Based on a number of airborne field programs, it is felt that the principal components of NO_y are generally well described in the UT largely because discrepancies between total measured NO_y and the individual components are often rather small. The minor species however are not known with high precision. Extensive measurements of NO_x have been made in concert with HO_x and other species to constrain the rate of ozone production in the UT. Based on a model constrained by extensive UT measurements, the loss rates of NO_x are found generally to exceed the production rates in large remote regions of the UT. Other measurements have been used successfully to quantify NO_x sources in

the UT, including lightning and aircraft emissions. The observations of NO_2 obtained from a new space borne instrument and of NO_x made on board commercial aircraft demonstrate the enormous potential of global scale measurements to lead to improved understanding of reactive nitrogen processes. A number of published studies (e.g. Emmons *et al.*, 1997) have compiled NO_x and NO_y data from one or more airborne campaigns and made these first-order climatologies available to the atmospheric sciences communities. A range of studies has used these data sets to demonstrate the skill of multidimensional models of the UT.

In order to improve our understanding using models, more extensive measurements in the UT are still needed. Specifically, more comprehensive measurements suitable to constrain NO_y production and loss processes would help define missing chemistry or unexpected uncertainties in accepted kinetic or photolytic rates. New measurements should also include minor NO_y species such as HO_2NO_2 and organic nitrates.

Laboratory measurements

The current state of our understanding of various reactions important in controlling the partitioning between NO_x and NO_y , and determining the abundance of the individual nitrogen oxide species were discussed. In addition, some key atmospheric reactions of species such as OH , HO_2 , and ClO that affect NO_x and NO_y abundance were discussed in the workshop; however, they are not highlighted below. Most of the key reactions coupling reactive nitrogen species within the NO_y reservoir have been measured in the laboratory and evaluated by the NASA Data Panel [NASA, 2000] and the IUPAC Panel [IUPAC]. The reactions discussed below and summarised in Table 2 are those considered to be of particular importance for further study

Table 2. Summary of priorities for future laboratory studies

Gas-phase processes
• $\text{OH} + \text{NO}_2 \rightarrow \text{Products}$ • Temperature dependence of $\text{O} + \text{NO}_2$ and $\text{NO} + \text{O}_3$ • Equilibrium of peroxyxynitrate species at atmospherically relevant temperatures • $\text{OH} + \text{HNO}_3$
Photochemical Processes
• $J(\text{HNO}_4)$ in the infrared region
Heterogeneous Processes
• The heterogeneous interaction of HNO_3 with water ice • Reactions that may convert HNO_3 back to NO_x • Surrogate surfaces for heterogeneous reactions

based on their role in NO_y photochemistry and the uncertainty associated with the reaction.

To validate atmospheric chemistry models in the UT/LS, it was suggested that rate constants should be determined to better than 15% accuracy. The general feeling was that this goal is presently difficult to achieve for many of the reactions involving NO_y species.

Gas phase processes

The most important gas-phase radical reactions for further study were determined based on their importance in the atmosphere or the large uncertainty associated with their rate constants. Those of high priority include the reactions of OH with NO_2 and HNO_3 , the reaction of O with NO_2 , the reaction of NO with O_3 , and the reactions that lead to the formation and destruction of RO_2NO_2 molecules. A major photochemical process in the atmosphere associated with large uncertainty is the overtone photolysis of HO_2NO_2 . These processes are very briefly discussed below.

• OH + $\text{NO}_2 \rightarrow$ Products (1)

Calculating ozone production is highly sensitive to the value of the rate coefficient for this reaction, which is one of the most investigated reactions. There is a wealth of data at 300K spanning more than four-orders-of-magnitude in pressure [NASA, 2000; IUPAC]. Currently, it is suspected that this reaction consists of two competing channels that result in HNO_3 and peroxyoxynitrous acid (HOONO) [Golden and Smith, 2000]. To date, HOONO has not been detected. The work of Hippler and co-workers on this reaction has provided evidence for the existence of an unstable intermediate thought to be HOONO (Hippler *et al.*, 2001). The O-O bond dissociation energy in HOONO is estimated to be $20.1 \text{ kcal mol}^{-1}$. Hippler *et al.* have fitted the available data at 300 and 400K as a function of pressure to this two-channel mechanism. The limiting high-pressure rate constants are consistent with the isotope scrambling experiments of Donahue *et al.* [2001] but are somewhat different from the interpretations of Golden and Smith (2000). The interpretation of Hippler *et al.* implies that one has to be careful in ascribing the measured OH temporal profile to either the sum of the two channels or to just one channel, because the assignment critically depends on the lifetime of HOONO relative to the OH observation time. The detec-

tion of HOONO , measurement of the rate coefficients for its formation, and interpretation of the existing data in terms of the two possible channels awaits further work.

• The temperature dependence of O + NO_2 and NO + O_3

The reaction $\text{O} + \text{NO}_2$, the rate limiting step in the NO_x -catalysed ozone destruction, was identified as an area of emerging confidence. Recent work, along with the some of the previous measurements, have better defined this rate coefficient and shown that it is ~15-20% higher at lower stratospheric temperatures than currently accepted values. The negative temperature dependence, albeit small, suggests that the reaction is not a simple abstraction reaction.

For the rate coefficient of the $\text{NO} + \text{O}_3$ reaction above 308K, the Arrhenius plot curves upwards such that a linear extrapolation of the high temperature data to lower temperatures becomes uncertain. This is further complicated by the possible differing rate coefficients for the reactions of the two spin-orbit states of NO with ozone and the changes in their relative abundance with temperature. Additional high quality data down to 200K, where the current uncertainty is approximately a factor of two [IUPAC, NASA, 2000], is desirable.

• The equilibrium of peroxyoxynitrate species at atmospherically relevant temperatures

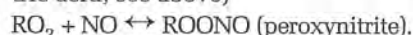
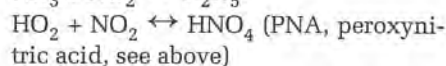
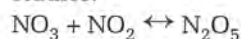
With the view that peroxyoxynitrates are atmospheric reservoirs of NO_2 , the equilibrium constants for reactions of the type $\text{RO}_2\text{NO}_2 \leftrightarrow \text{RO}_2 + \text{NO}_2$ are needed over a large temperature range to accurately predict the atmospheric fate of RO_2NO_2 . RO_2 radicals may be both alkylperoxy and acylperoxy radicals. Both forward and reverse reactions involving CH_3O_2 , $\text{C}_2\text{H}_5\text{O}_2$, and $\text{CH}_3\text{C(O)O}_2$ and the thermal decompositions of $\text{CH}_3\text{C(O)O}_2\text{NO}_2$ and $\text{C}_2\text{H}_5\text{C(O)O}_2\text{NO}_2$ have been studied. Future studies should extend the temperature range for forward and reverse reactions, obtain absolute rate constants for these reactions, and obtain kinetic data as a function of pressure to separate the low- and high-pressure rate constants.

• OH + HNO_3

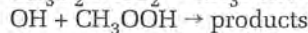
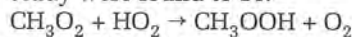
This reaction has recently been extensively studied by Brown *et al.* (1999a,b; 2001) and has been assessed by the NASA panel. However, an independent confirmation of the Brown *et al.* results is

warranted. The rate coefficient, which increases with decreasing temperature, is pressure dependent. The only products are H_2O and NO_3 , even though reaction proceeds via the formation of a complex, $\text{HNO}_3 \cdot \text{OH}$, that is long enough lived to make the rate coefficient pressure and bath-gas dependent at low temperatures.

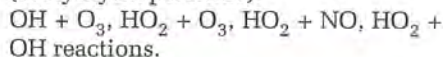
The following reactions also were found to have high priority for future studies:



Other important atmospheric reactions that indirectly impact the NO_x to NO_y ratio and the abundance of the NO_x and NO_y species, and require further study were found to be:



Formation and fate of $\text{CH}_3\text{C(O)OOH}$ (acetylhydroperoxide).



Photochemical processes

• J(HNO_4) in the infrared region

Overtone photolysis of HO_2NO_2 was highlighted in this workshop. Even though the photodissociation cross section in the near infrared spectral range may be quite small, the rate of this process may be significant owing to the large actinic flux in this spectral region. The need for further laboratory work on the photochemistry of HO_2NO_2 to reduce the uncertainty in the UV and IR absorption cross sections was stressed. A new value for the standard enthalpy of formation of HO_2NO_2 of $-52.7 \pm 8 \text{ kJ mol}^{-1}$, compared to the old value of $-57.24 \text{ kJ mol}^{-1}$, shifts the dissociation threshold from 8787.2 to 8407.4 cm^{-1} . However, this change is not sufficient to explain the photochemical action spectrum obtained at -20°C that showed photodissociation activity around 7000 cm^{-1} (Wennberg *et al.*, 1999). However, this results needs confirmation.

Other photochemical processes that need further study were identified to be the photochemistry of PAN and photolysis quantum yield of CH_2O .

Heterogeneous processes

• The heterogeneous interaction of HNO_3 with water ice

The gas phase lifetime of HNO_3 strongly depends upon the coefficient, γ for the uptake on cirrus clouds. The value of γ is constant at 0.3 in the range 170

to 195K and decreases with temperature between 195K and 240K [IUPAC, NASA, 2000]. Certain uptake experiments of HNO_3 on H_2O -ice substrates simulating cirrus clouds have resulted in unlimited uptake up to a temperature of 210K whereas other experiments resulted in significant saturation of the HNO_3 uptake below 210K (Zondlo *et al.*, 2000). Currently, the temperature above which saturation of the ice surface by HNO_3 occurs is uncertain.

Several workers have proposed a significant decrease of the H_2O evaporation rate from ice upon HNO_3 uptake (Livingston and George, 1998; Warshawsky *et al.*, 1999) and thereby extending the lifetime of cirrus clouds. In contrast, such an effect was not observed in another study (Beirmann *et al.*, 1998). A decrease of the H_2O evaporation rate due to the presence of adsorbed HBr [Hudson *et al.*, 2001] and HCl on ice (Delval *et al.*, 2001) has also been obtained. No definite assessment of the role of adsorbed HNO_3 , HCl, and HBr on ice may be given at this time with regards to the resulting lifetime of atmospheric ice particles. Quantification of the changes in H_2O evaporation rates due to HNO_3 uptake as a function of temperature is needed. In addition, the nature of the coating with respect to the evaporation rate of H_2O on ice needs to be carefully assessed in view of several thermodynamically stable or metastable structures.

• Reactions that may convert HNO_3 back to NO_x

A few reactions were considered that might convert HNO_3 back to NO_x and, thereby, alter the NO_x to NO_y ratio in the UT. They include the reaction of CH_2O with HNO_3 to give $\text{HCOOH} + \text{HONO}$ in sulphuric acid aerosols and the possible reduction of HNO_3 to NO_2 in droplets. Even though the first reaction has been studied a few times, more quantitative information is needed to assess its role in the atmosphere.

• Surrogate surfaces for heterogeneous reactions

In general, laboratory studies of heterogeneous reactions have been carried out using surrogates for atmospheric substrates. It has recently become clear that structural and chemical features of the condensed phase can control both the kinetics of the uptake and the identity of the products. Furthermore, the gas uptake itself may alter the surface and the bulk substrate. Substrates of interest include ice, soot, and mineral dust. The soot generated in the laboratory may not represent all of the features of atmo-

spheric soot. Laboratory studies alone are not sufficient to derive the properties of environmental soot particles; field measurements are badly needed. In the absence of field characterisation, soot with differing properties have been generated by adjusting the combustion conditions from fuel lean to fuel rich.

Another drawback of using the surrogates is the correction for the porosity of the substrate. In a number of cases the correction for porosity has been included without experimental verification of whether or not such an application of pore diffusion theory was justified. For "sticky" molecules, whose desorption rate constant is significantly slower than the rate of surface reaction, it is expected that pore diffusion will not be important because the reaction takes place before the molecule has a chance to explore the internal surface. Conversely, in the case of a fast desorption rate, one needs to correct the observed rate of uptake because of an enhanced gas-surface collision rate.

Many other issues were briefly discussed but they are not included in detail here. The discussed topics include coating of combustion aerosol (soot) particles by sulphuric acid; producing internal and external mixtures of aerosols and their surrogates for laboratory studies [Jacobson, 2001]; and dealing with mineral dust particles in the context of laboratory studies, and identifying the best model substrates for laboratory studies.

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Variability of the Tropical Tropopause

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Introduction

It is well known that much of the mass imported into the stratosphere rises through the tropical tropopause (TT). The focus on the TT during the recent SPARC Tropopause Workshop (see Haynes and Shepherd, 2001) reflects increased interest in the need to understand the details of stratosphere-troposphere exchange (STE) processes within the tropics on a variety of timescales. In particular, the study of the relationship between STE and tropical convection has developed renewed momentum given the longer term *in situ*, reanalysis, and satellite data now available to document this variability.

Despite the mounting recognition that the TT should be considered as a vertical layer rather than a sudden transition between the troposphere and the stratosphere (Highwood and Hoskins, 1998; Gettelman *et al.*, 2001a), the TT is often conveniently defined either as an abrupt change in the lapse rate or as the so-called “cold point” of an atmospheric temperature sounding. This approach greatly simplifies the quantification of the climatology and variability of the TT using observational data.

In this note we attempt to consolidate some recent observations concerning the large-scale relationships between the TT, tropical convection, and stratospheric conditions on various time scales. For the bulk of the results shown here, the TT was defined according to either the WMO lapse-rate definition or the cold point tropopause (CPT).

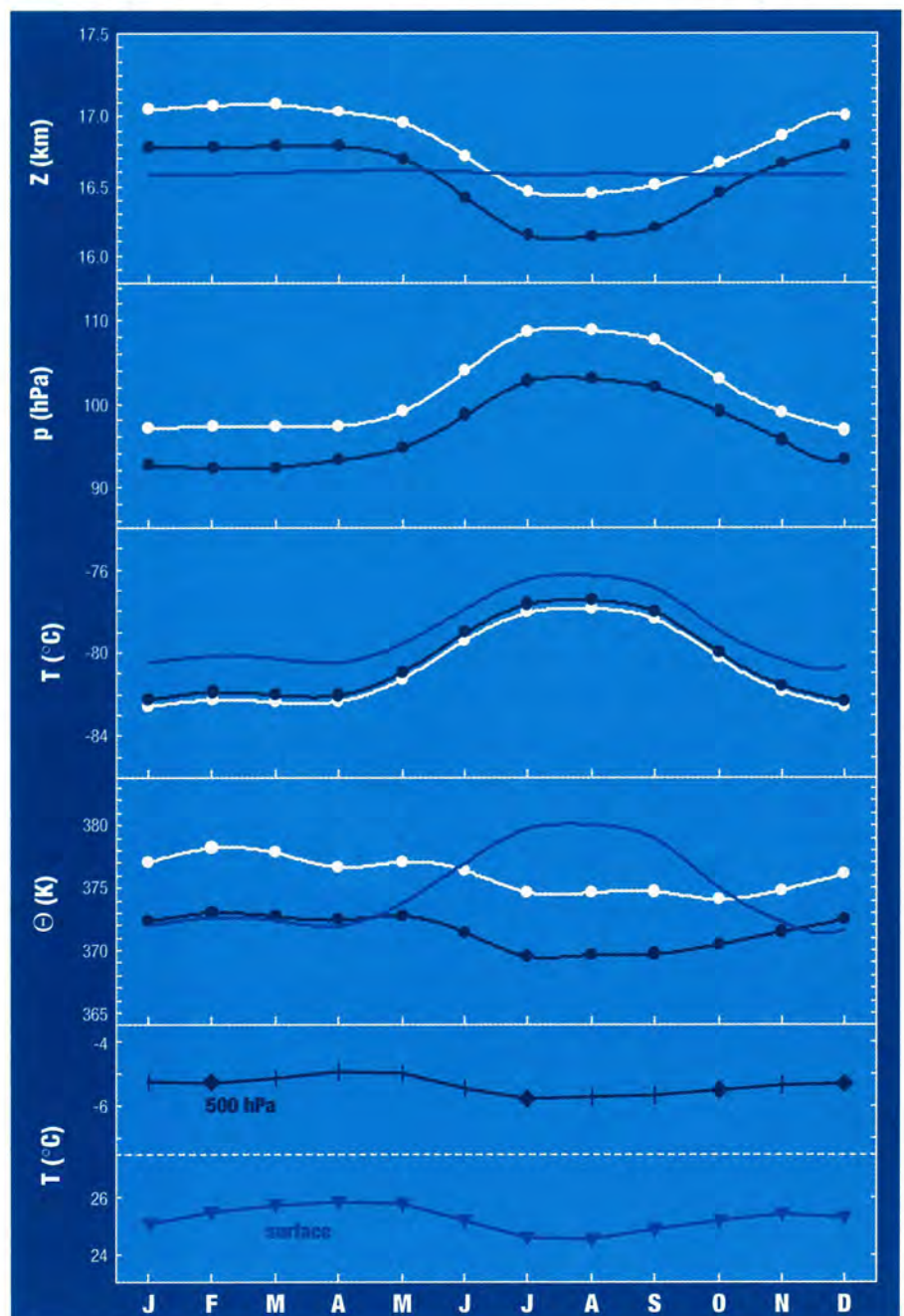
Seasonal cycle

Climatological depictions of the TT based on radiosonde data (Makhover, 1979; Seidel *et al.* 2001) and on reanalyses (Hoinka, 1999; Randel *et al.*, 2000) reveal a primarily zonally symmetric structure with maximum height and minimum temperature near the equator. Animated graphic loops of the seasonal variations in TT temperature, pressure, height and saturation mixing ratio, derived from radiosonde data (Seidel *et al.*, 2001), can be viewed at

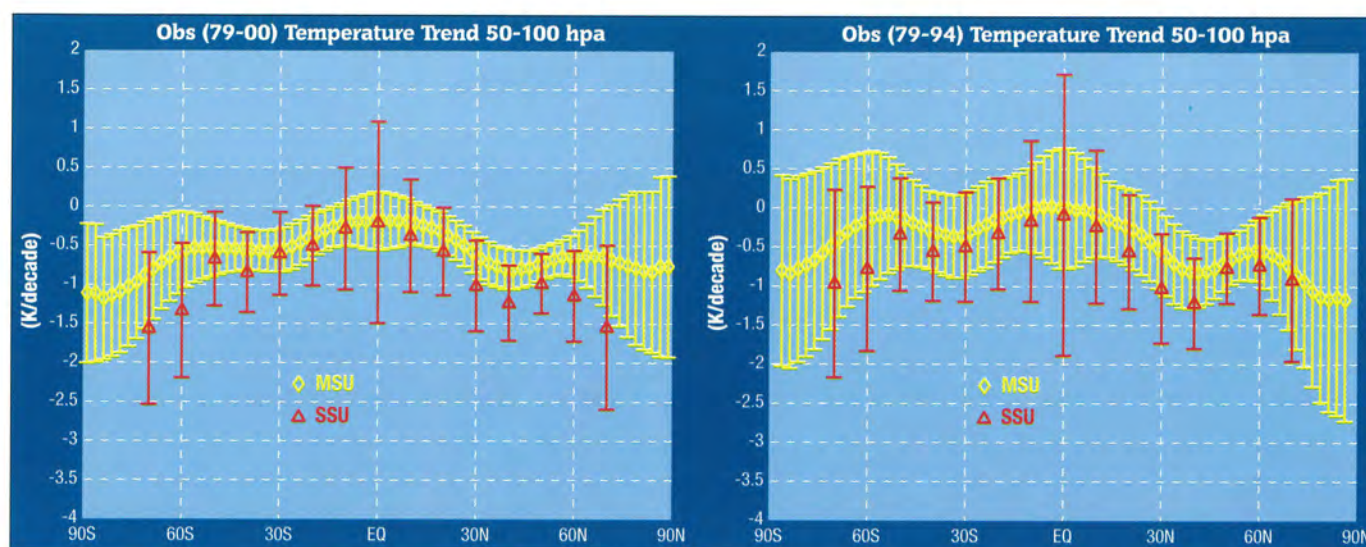
the SPARC Data Center at: http://www.sparc.sunysb.edu/html/noaa/noaa_trop.html. A striking feature of these plots is that the TT is highest, cold est, and at its lowest pressure throughout the tropics during Northern

Hemisphere (NH) winter, with the opposite extremes exhibited in NH summer. As depicted in Figure 1, this seasonality is evident in the equatorial region (10°N-10°S), where the amplitude of the zonal-mean annual cycles of the

Figure 1. Climatological annual cycle of equatorial tropopause characteristics based on radiosonde data averaged from 10°N to 10°S for 1961-1990. The temperature, height, pressure, and potential temperature are shown for the cold point (white dots), lapse-rate tropopause (black dots), and the 100 hPa level (thin blue curves). Temperatures at the surface and 500 hPa are also shown for comparison. [Adapted from Seidel *et al.* (2001)].

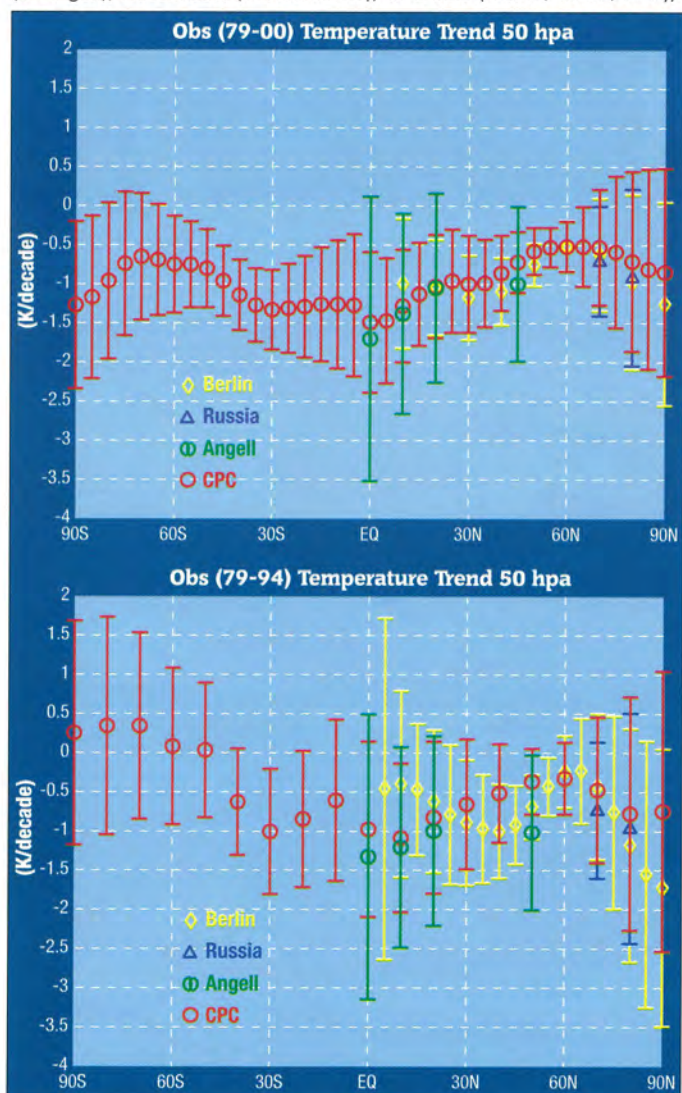


An Update of Stratospheric Temperature Trends

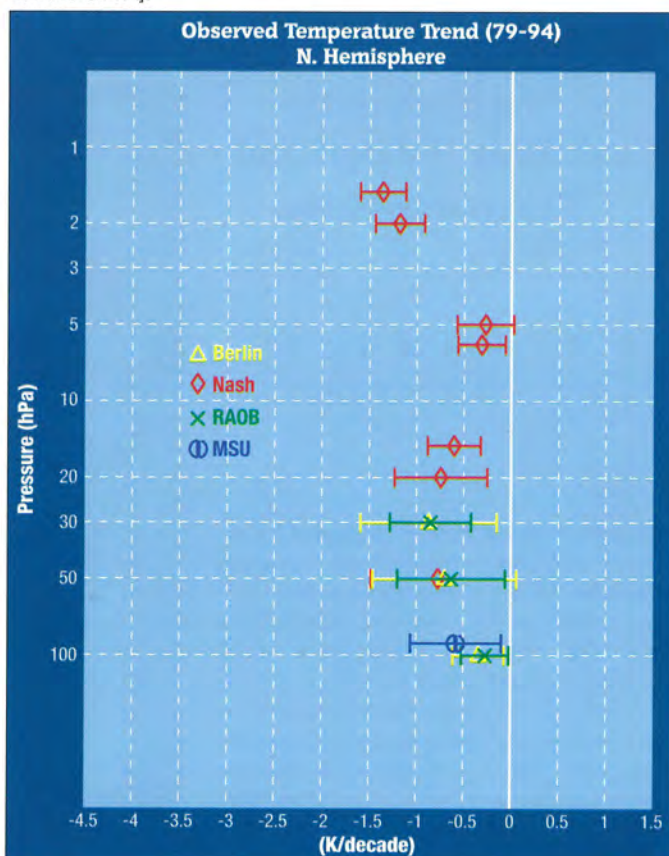


▲ **Figure 1** - Zonal, annual-mean temperature trends (K/decade) in the ~50-100 hPa region, as derived from the MSU and SSU 15X satellite observations. Left panel is trends for the 1979-2000 (the SSU data extends only through 1998) period while the right panel is for the 1979-1994 period. Also shown in both panels are the corresponding 2-sigma level uncertainties. [Courtesy: MSU data (J. Christy); SSU data (J. Nash); assembled by SPARC-STTA].

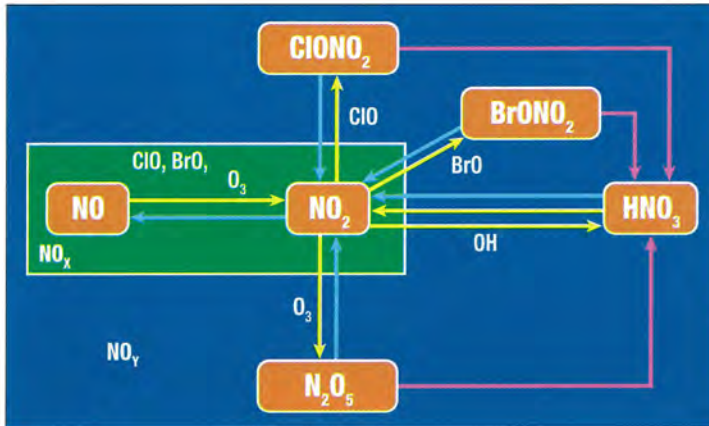
▼ **Figure 2** - Same as Figure 1, except for non-satellite data (top for the 1979-2000 period, bottom for the 1979-1994). The radiosonde data sets consist of Angell, Berlin and Russia, while CPC comprises an analyses data set. [Courtesy: Berlin data (K. Labitzke and B. Naujokat); Angell data (J. Angell); Russia data (Y. Koshelkov); CPC data (NOAA/ NCEP/ CPC); assembled by SPARC-STTA].



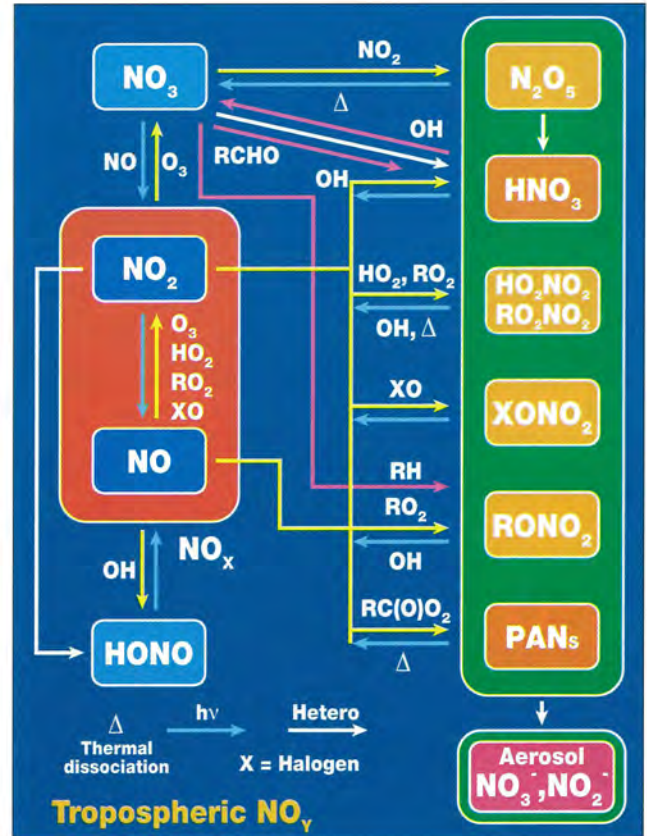
▼ **Figure 3** - Vertical profile of the annual-mean Northern Hemisphere trends from various satellite and radiosonde data sets for the 1979-1994 period. Note that the Nash (same as "SSU") and MSU data, as plotted here, denote the peak altitude of emission whereas the emission comes from a broad range of altitude surrounding the peak emission layer. [Courtesy: UKRAOB data (D. Parker); assembled by SPARC-STTA].



Report on the Nitrogen Oxides in the Lower Stratosphere and Upper Troposphere Workshop, University of Heidelberg, Heidelberg, Germany, 19-22 March 2001



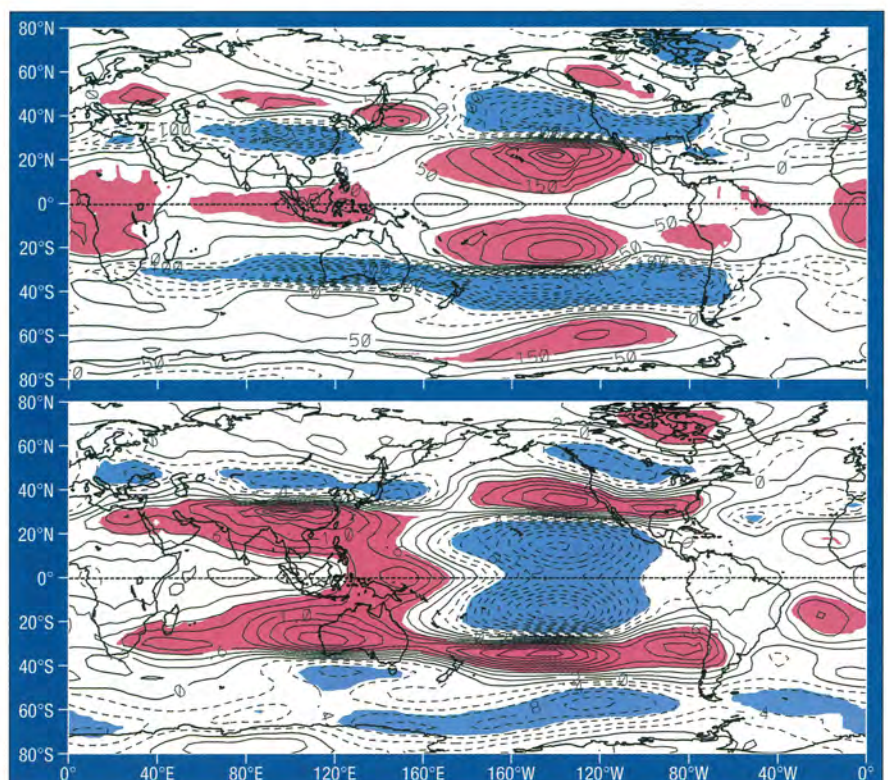
▲ **Figure 1**
Schematic of reactive processes in the stratospheric NO_y reservoir: gas-phase (yellow), photolytic (blue), and heterogeneous (pink). [From Fahey and Ravishankara (1999)].

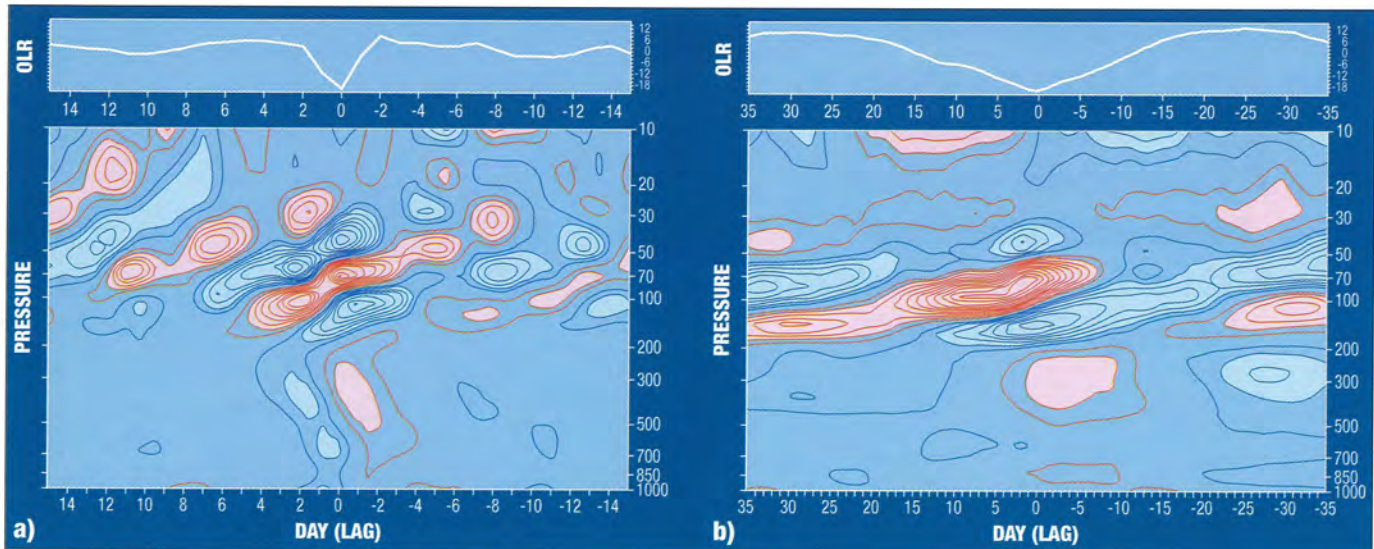


► **Figure 3**
Schematic of tropospheric NO_y species and their interconversion processes. [Courtesy of B. Ridley].

Variability of the Tropical Tropopause

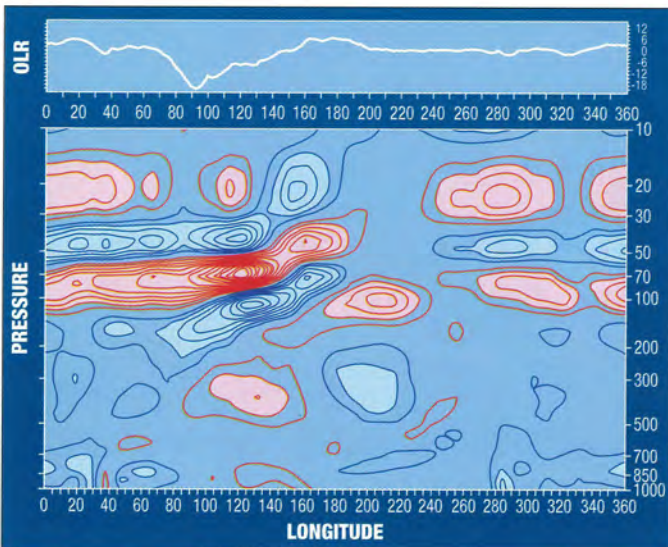
► **Figure 2** - Maps of (a) tropopause height and (b) tropopause temperature regressed onto Niño 3.4 sea surface temperature (SST) anomalies, using monthly data from 1958-1999. Values are plotted for a $+2.0^\circ\text{C}$ perturbation in SST. Contour interval is 50 m in (a), and 0.2°C in (b). Positive (negative) perturbations shown as solid (dashed) contours. Red (blue) shading denotes statistically significant positive (negative) perturbations. [Adapted from Kiladis *et al.* (2001)].





▲ Figure 3

Outgoing longwave radiation (OLR) and time-height temperature perturbations from Singapore radiosonde data regressed against (a) a Kelvin wave index (see text) for December-February 1979-1999 and (b) a Madden-Julian Oscillation index. The perturbations have been scaled to a -20 Wm^{-2} anomaly in OLR at the station at simultaneous lag (Day 0). Lags are in days, with time going from right to left, and data have been plotted on pressure surfaces. Contour interval is 0.1°C . Red (blue) shading denotes positive (negative) perturbations.



◀ Figure 4

As in Figure 3, but a longitude-pressure plot of temperature along the equator on Day 0 from December-February 1979-93 ECMWF reanalysis data versus the MJO index. Contour interval is 0.1°C .



The global Atmosphere Watch Monitoring Components

◀ Figure 2

The present network of the 22 GAW Global stations.

European Research in the Stratosphere, 1996-2000: Advances in our Understanding of the Ozone Layer during THESEO

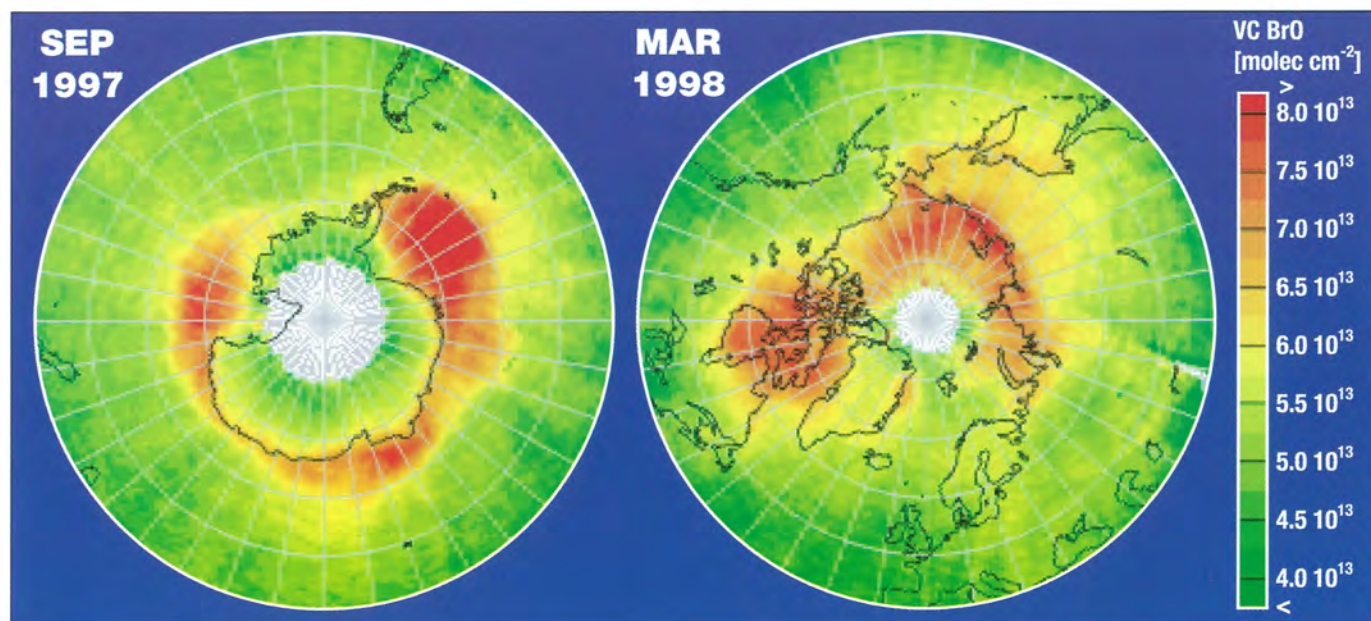


► Figure 1

"The contrail of the Geophysica aircraft flying at 17 km altitude seen from the DLR Falcon aircraft (at 12 km) on the way to the tropical cyclone Davina on 9 March 1999 during the APE-THESEO experiment. Bands of thin cirrus clouds are visible, belonging to the extended spiral arms of the cyclone. The two aircraft fly in tandem, with the Falcon acting as a pathfinder (by using its aerosol lidar) to steer the Geophysica into ultrathin tropopause clouds which were invisible to the Geophysica pilot."

IV

Reports on the IAMAS General Assembly, 10-18 July 2001, Innsbruck, Austria



▲ Figure 1

The abundance of tropospheric BrO measured from the GOME satellite. Such measurements of reactive species from space highlight the capability of satellites in providing valuable atmospheric data.

tropopause temperature, height, and pressure are approximately 5K, 600 m, and 10 hPa, respectively. The annual variations in equatorial mid-tropospheric (500 hPa) and surface temperature are considerably smaller (about 1K) and show evidence of a semi-annual signal, suggesting that the seasonality of the TT is not driven by thermal conditions in the underlying troposphere. Seasonal variations in wave-driving, which is strongest in NH winter and spring, are more likely responsible (Holton *et al.*, 1995; Seidel *et al.*, 2001).

Figure 1 also depicts the systematic differences between the TT defined on the basis of the lapse rate and the cold point. Although the temperature of the two levels is approximately the same, the cold point is located about 300 m higher and its potential temperature is higher by about 5K than that of the lapse rate tropopause. The 100 hPa level, which has in the past been used as a convenient surrogate for the TT, does not share the same seasonal variability in height as the TT. With new TT data sets based on reanalysis data (Randel *et al.*, 2000) and radiosonde data (Seidel *et al.*, 2001) now available via the SPARC Data Center, we advise against the use of the 100 hPa level in tropopause studies.

Despite its zonal symmetry, there are marked asymmetries about the equator in the mean seasonal cycle, with the tropopause being higher, colder and at lower pressure in the Northern Hemisphere (NH) than in the Southern Hemisphere (SH) during NH winter. In NH summer, the tropopause is higher and at lower pressure in the SH, but it remains colder in the NH.

Interannual variability

Several recent studies point to the El Niño/Southern Oscillation (ENSO) phenomenon as being a significant factor in interannual variability of the TT, along with the stratospheric QBO (e.g. Randel *et al.*, 2000; Zhou *et al.*, 2001a). Other, more subtle effects include solar forcing (van Loon and Labitzke, 1999) and changes in the residual circulation due to interannual variations in extratropical wave driving (e.g. Salby *et al.*, 2000). To demonstrate the impact of ENSO, we utilise the "Niño 3.4" sea surface temperature (SST) index, obtained by monthly averaging the SST between 5°N–5°S, 170°W–120°W, and subtracting from these values a 1961–1990 climatology. As is well-known, equatorial convection is strongly related to

the Niño 3.4 index. During warm events when SST is anomalously high in the central tropical Pacific, there is enhanced convection over that sector east of 140°E, with maximum amplitude just south of the equator near the dateline (Kiladis *et al.*, 2001). A region of suppressed convection is then seen over the eastern Indian Ocean and Indonesia, extending into the South Pacific Convergence Zone. Nearly opposite conditions are observed during cold events.

A global view of the response of the tropopause to tropical Pacific SST variations, derived from NCEP/NCAR reanalysis, is shown in **Figure 2** (p. II). These maps show the simultaneous regression of tropopause height (Z_{trop}), and temperature (T_{trop}) scaled to a +2.0°C perturbation in Niño 3.4 SST anomalies for the period from June 1958 through May 1999. Throughout most of the tropics between about 20°N and 20°S, a higher (lower) than normal tropopause accompanies warm (cold) events in Niño 3.4 SST, confirming earlier work of Gage and Reid (1987). Pictures of T_{trop} similar to that shown in **Figure 2a** for Z_{trop} were obtained for the tropical belt by Randel *et al.* (2000), Zhou *et al.* (2001a), and Gettelman *et al.* (2001b). This signal reaches its highest positive amplitude in the subtropical Pacific at 20°N and 20°S, and around 140°W longitude, with nearly equal amplitude negative perturbations of Z_{trop} poleward of these locations at 40°N and 40°S. To the west, the negative Z_{trop} regions shift equatorward to 30°N and 30°S, extending from the Atlantic, across the Indian sector to eastern Asia and Australia.

The T_{trop} pattern corresponding to **Figure 2a** is shown in **Figure 2b**. While there is a strong zonally-symmetric tropical Z_{trop} signal related to ENSO, T_{trop} has a much more zonally-varying structure at low latitudes. In fact, as **Figure 2b** suggests, the T_{trop} difference between Tahiti and Darwin is highly correlated with the more standard Southern Oscillation Index based on surface pressure at those locations (Randel *et al.*, 2000). A comparison of **Figures 2a** and **2b** shows that almost everywhere in the extratropics where the ENSO signal is manifested, there is a strong negative correlation between Z_{trop} and T_{trop} , such that when the tropopause is high (low) it is also cold (warm). This is also true throughout the subtropical and equatorial central and eastern Pacific. However, despite the higher

than normal tropopause over the western Pacific, T_{trop} there is actually anomalously warm, as was observed in the radiosonde data by Reid *et al.* (1989) and Kiladis *et al.* (2001). The geographical extent of the region where the tropopause is high and also warm corresponds closely to the region of suppressed convection centred on Indonesia. This suggests that the origin of this tropopause warming might be increased upwelled longwave radiation from the surface, since this area is less cloudy than normal during ENSO warm phases. This mechanism is also consistent with the results of Thuburn and Craig (2000), who imply a sensitivity in the height of the tropopause to the amount of upwelling longwave radiation.

In **Figure 2a**, local maxima in tropical geopotential height anomalies occur in the subtropical Pacific of both hemispheres at 140°W, the same locations as the minima in T_{trop} in **Figure 2b**. This is the sector where the anomalous convection over the Pacific sector is associated with the most pronounced enhancement of the local Hadley circulation, leading to a spin-up of the Pacific subtropical anticyclones. This signal also appears as anticyclonic potential vorticity (PV) anomalies maximised close to tropopause level (not shown), which are a response to enhanced upper tropospheric mass outflow from the anomalous equatorial convection. As discussed by Hoskins *et al.* (1985), anticyclonic PV perturbations in the upper troposphere (UT) lead to an upward displacement in the tropopause, and require cold temperature anomalies above and warm temperature anomalies below them to maintain hydrostatic balance, as seen in vertical cross sections of temperature perturbations at these longitudes (Kiladis *et al.*, 2001).

Hydrostatic and PV arguments also provide a convenient framework for considering the tropopause perturbations closer to the equator. Highwood and Hoskins (1998) studied the large scale response of the tropical tropopause to convective heating through the use of a dry primitive equations model, in which a heat source was imposed along the equator. This produced subtropical anticyclonic gyres to the west and a Kelvin response with westerlies to the east of the heat source at upper levels. The vertical structure featured increased (decreased) pressure in the upper (lower) troposphere, with subsidence and warming of the middle and lower troposphere, and

upward motion and cooling extending through the UT, the tropopause region, and into the lower stratosphere (LS), as is observed.

It seems evident that, on ENSO time scales at least, the large scale pattern of perturbations in the tropopause at most tropical locations can be explained as a radiative-dynamical response to diabatic forcing by the anomalous SST distribution. Given the results of Salby *et al.* (2000), however, we cannot discount the possibility that “downward control” principles may be of some importance to the details of the distribution. Further study of the fine structure of these perturbations may help in better quantifying the relative importance of diabatic and dynamical effects on the tropopause.

Intraseasonal variability

It has long been known that the TT responds significantly to the passage of equatorially-trapped disturbances in the LS. Recent work has shown that large perturbations in temperature in the LS and at the tropopause level are associated with convectively coupled waves propagating zonally along the equator (Wheeler *et al.*, 2000; Kiladis *et al.*, 2001; Straub and Kiladis, 2001). **Figure 3** (p. III) shows time-height cross sections of temperature perturbations from radiosonde data at Singapore associated with the passage of two important convectively-coupled equatorial disturbances, the Madden-Julian Oscillation (MJO) and Kelvin waves. **Figure 3b** was produced by regressing Singapore temperature data against an index of the MJO obtained by combining the first two EOFs of 120-day high-pass filtered outgoing longwave radiation (OLR) between 20°N and 20°S. **Figure 3a** was obtained in a similar way except by using space-time filtering OLR to retain only fluctuations in the Kelvin wave band (see Wheeler and Kiladis, 1999). Both plots are scaled to a -20 Wm^{-2} perturbation in OLR on Day 0, which is a typical value of a perturbation associated with these disturbances as they pass by the station.

In both disturbances, the largest temperature perturbations are observed around the TT, extending vertically in wavelike perturbations into the LS. For the Kelvin wave (**Figure 3a**), the largest negative temperature perturbation at the TT (near 100 hPa at this location) occurs 2 days prior to the passage of the convection, with a peak positive perturbation occurring two

days following the convective signal. For the MJO (**Figure 3b**), the sequence is similar but the perturbations are even larger, and the peak TT perturbations occur more than a week prior to and following the convective peak. In fact, the TT temperature perturbation is near zero when convection is maximised at Singapore for both disturbances.

This vertical structure is consistent with the forcing of vertically propagating energy by an upper tropospheric energy source provided by the convective heating associated with the disturbance. For an upper tropospheric source moving eastward along the equator, temperature phase lines should tilt upward and eastward above the source, as in a vertically propagating gravity wave (Andrews *et al.*, 1987). A zonal cross section of the MJO temperature perturbation along the equator (**Figure 4**, p. III), derived from ECMWF reanalysis data, is consistent with this implied structure. The Kelvin wave shows similar structure although at a smaller spatial scale (Wheeler *et al.*, 2000). In **Figure 4** upward and eastward tilting phase lines are seen well to the east of MJO convection, leading by more than 20 degrees of longitude. The time evolution of this signal (not shown) reveals that the entire perturbation pattern shifts eastward at between $5\text{--}10 \text{ ms}^{-1}$ for the MJO, while in the Kelvin wave, the phase speed is somewhat faster, at around 15 ms^{-1} . Thus TT perturbations, as well as any potential STE mechanisms that might be dependent on the vertical motion associated with these waves, while substantial, are not collocated with local convection. Fujiwara *et al.* (1998) found convincing evidence of the STE of ozone associated with Kelvin waves and possibly the MJO, inferred from radiosonde and ozonesonde data over Indonesia. For the Kelvin wave the relationship was such that maximum upper tropospheric ozone concentrations occurred during periods of anomalously low tropopause and downward propagating warm temperature perturbations, implying downward transport from the stratosphere across the tropopause due to the dynamical structure of the wave. Temperature fluctuations in the TT region due to Kelvin waves have also been implicated in cirrus cloud formation at that level (Boehm and Verlinde, 2000).

While the largest TT perturbations are generally observed at some distance

away from the deepest convection coupled to equatorial waves, there is ample evidence that local convection can penetrate through the TT into the LS (Gettelman *et al.*, 2001b). The precise nature of the impact of this higher frequency convection on the TT and in STE, and in particular its role in the dehydration of the stratosphere, is an area of active research (Sherwood, 2001).

Quasi-Biennial variations

Distinct quasi-biennial oscillations (QBO) of the TT have long been recognised (Angell and Korshover, 1974; Reid and Gage, 1985) and reveal a certain level of control over the TT by the equatorial stratosphere. Randel *et al.* (2000) find that the QBO explains about 12% of the interannual variance in TT temperature and height, with higher equatorial tropopause temperatures associated with westerly QBO winds in the equatorial stratosphere. In the subtropics, TT QBO temperature variations are of opposite sign to equatorial signal.

Using ECMWF reanalysis data, Zhou *et al.* (2001a) clearly depict the zonally symmetric structure of the QBO signal in the cold-point tropopause (**Figure 5**, p. 21). Here the zonal wind shear at 50 hPa over Singapore is taken as an index of the QBO in equatorial lower stratospheric winds. Zhou *et al.* (2001a) find that the TT anomalies lag the QBO by about six months, suggesting a downward-propagating signal consistent with the dynamics of the QBO.

Long-term changes

The long-term variability of the TT is not well understood. Suggestions of a solar cycle influence on the TT have been made (Reid and Gage, 1985; van Loon and Labitzke, 1999), but these signals are much less significant than the temperature signals in the LS observed to be associated with the solar cycle. Large volcanic eruptions warm the stratosphere, and these also may influence TT characteristics, but observational evidence is limited. Randel *et al.* (2000) identify a warming and lowering of the TT following the 1982 eruption of El Chichón, but the larger 1991 Pinatubo eruption appears to have had a less notable influence.

Figure 6 shows temperature variations from radiosonde data in the 15°S to 15°N latitude band at the TT and at three other levels, along with tropopause

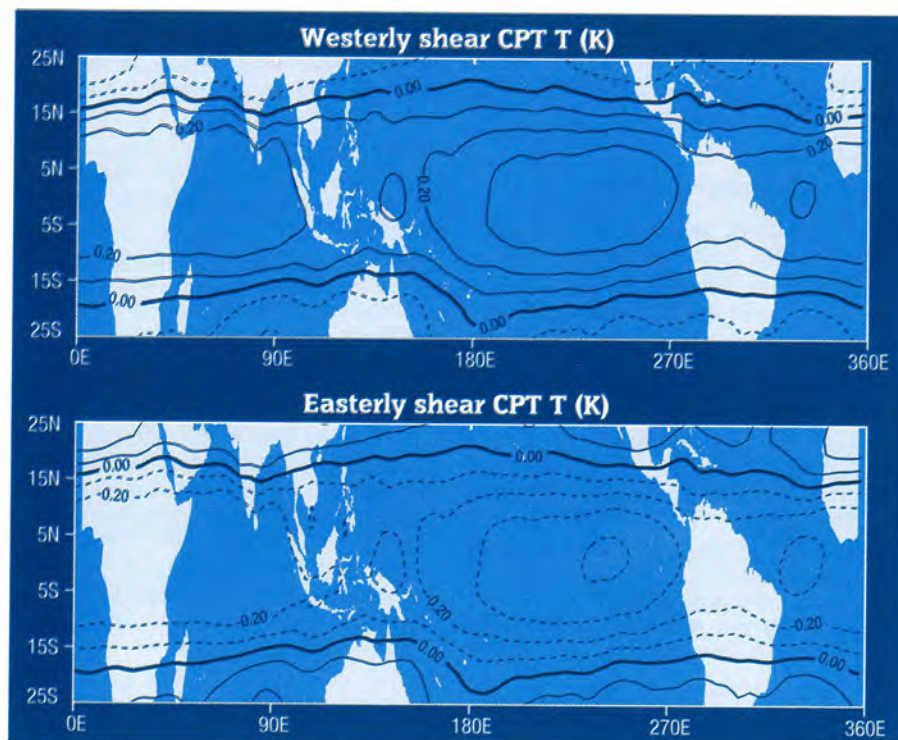


Figure 5. Composite analyses of CPT temperatures under westerly and easterly stratospheric zonal wind shear conditions. Winds at 40 and 70 hPa at Singapore were used to estimate 50 hPa wind shear. Note that the shears lead the CPT temperatures by six months. Contour interval is 0.1°C . [Adapted from Zhou *et al.* (2001a)].

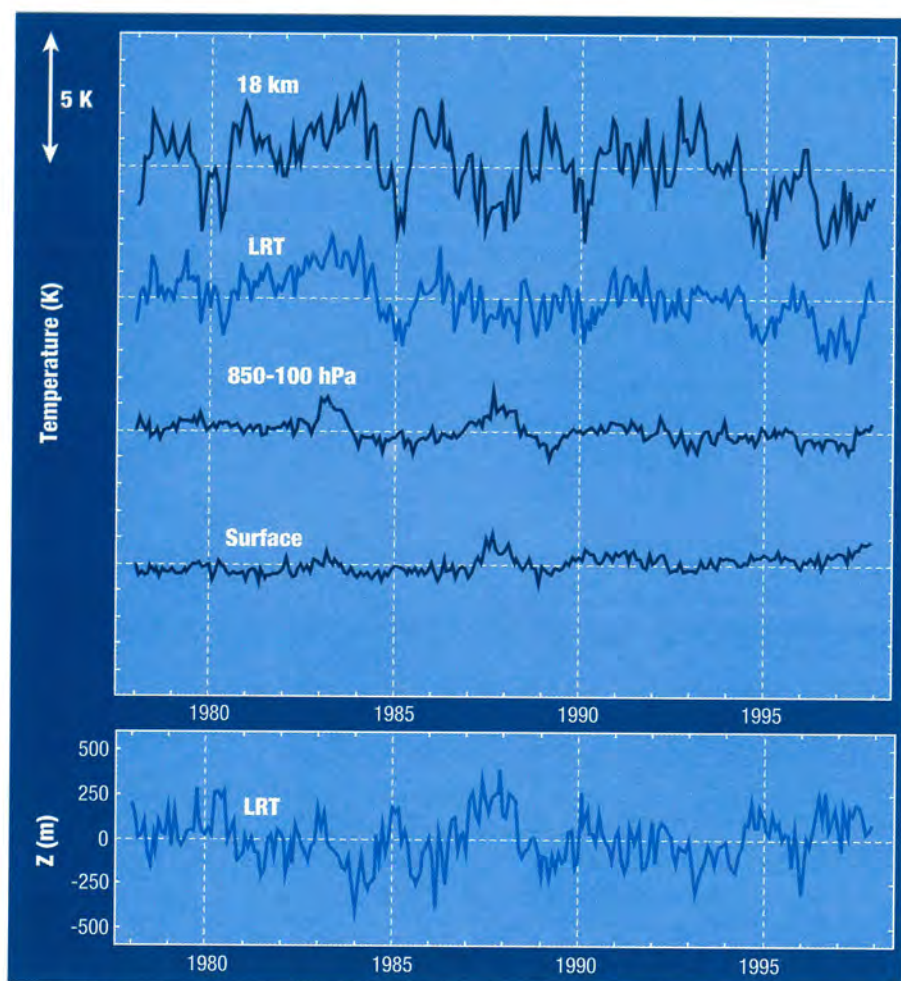


Figure 6. Monthly anomalies of temperature at 18 km, at the lapse rate tropopause (LRT), in the 850-100 hPa layer, and at the surface, averaged from 15°N - 15°S , for 1978-1997 based on radiosonde observations. [Adapted from Seidel *et al.* (2001)].

height variations. Compared with the troposphere and surface, the TT temperature shows much larger inter-annual variability, although not quite as much as the LS (18 km level). The variations in TT temperature are also much more highly correlated with LS temperature ($r = 0.74$) than with surface ($r = -0.01$) or tropospheric ($r = 0.13$) temperature. On the other hand, the height of the tropopause covaries both with stratospheric temperature ($r = -0.80$) and tropospheric temperature ($r = 0.49$).

A number of recent studies have attempted to evaluate multi-decadal trends in the TT, motivated largely by the need to determine whether recently identified increases in stratospheric water vapour (SPARC, 2000; Rosenlof *et al.*, 2001) can be explained in terms of changes in the TT. Unfortunately, existing data sets are not well suited to TT trend detection. Reanalysis data are severely affected by changes in the available observational data, particularly the introduction of satellite observations in 1979 (Randel *et al.*, 2000). Radiosonde data are affected by instrument changes that compromise the long-term homogeneity of the observations (Seidel *et al.*, 2001). Nevertheless, three recent studies (Randel *et al.*, 2000, Seidel *et al.*, 2001, Zhou *et al.*, 2001b) find suggestions of a cooling of the TT since the 1970's, as indicated in Figure 6. The downward trend in tropopause temperature, of about 0.5K/decade , is consistent with the cooling of the stratosphere (Ramaswamy *et al.*, 2001) and opposes the marked surface warming, of about 0.15K/decade , in this region. The TT cooling is associated with a decrease in stratospheric entry-level saturation water vapour mixing ratio and so does not explain the observed water vapour trends. However, other changes, perhaps in the areal extent and frequency of STE or in the magnitude of the mass flux through the TT, may play important roles and certainly merit further study.

Challenges

This brief overview highlights the complex nature of the TT region and various aspects of TT variability over a wide range of timescales. This variability appears to manifest elements of tropospheric changes, such as those related to the location and timing of convection on ENSO and intraseasonal time scales, as well as stratospheric variations such as the QBO and volcanic

influence. The zonal symmetry along with asymmetries about the equator of the annual cycle seem to be driven by stratospheric processes, and recent downward trends in TT temperature are consistent with stratospheric cooling. Furthering our understanding of the nature of TT variability will require advances in both observations and theory.

Small time and space scale variations in the TT have received less attention to date, probably due to the lack of data with adequate resolution. Such observations are needed to better delineate the diurnal and day-to-day variations in the tropopause, which are not well documented. A better understanding of possible TT variations on small spatial scales would shed light on the nature of stratosphere-troposphere exchange, particularly of water vapour. The current radiosonde network, which is particularly sparse in the tropics, cannot resolve variations with length scales of order 1000 km or less.

Increased spatial coverage by radiosondes and other *in situ* sensors will be necessary to isolate the finer structure of TT variations. The detection of TT trends will rely primarily on continuous, high quality, long-term sounding data. Satellite observations with improved vertical resolution near the TT would also help to fill these needs, but current sounding techniques do not yet provide the resolution afforded by radiosondes. Coordinated meteorological and chemical measurements are also needed to enable progress in STE research.

The development of models with complete coupling of the stratosphere and troposphere is progressing. Given the observed associations of the TT with tropical waves, future models will have to display reasonably adequate spectra of tropical disturbances, down to the high frequency gravity waves responsible in part for driving the QBO. This in turn will require continued improvements in the parameterisation of radiation and convection. Further advances in the coupling of interactive chemistry will be necessary to simulate details of STE processes, and to assess the extent of the impacts of various global change scenarios. While great progress has been made in this type of model development over the past few years, as stated by Haynes and Shepherd (2001), this subfield is "still in its infancy". The successful melding of the observational, theoretic-

cal, and modelling aspects of the TT and associated STE processes poses a particularly daunting cross-disciplinary challenge.

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Report on the Summer Monsoon Water Vapour Workshop

Boulder, USA, 11 October 2001

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A mini-workshop took place at the NCAR to review what is known about the structure of the Northern Hemisphere (NH) summer monsoons over Asia and North America, and to discuss the role of these circulation systems in the seasonal and long-term evolution of water vapour in the upper troposphere and lower stratosphere (UT/LS). The workshop discussion included a comprehensive overview of existing and planned observations that provide information on monsoon circulations and transport in the UT/LS. These observations consist of ground-based, airborne, and satellite measurements of water vapour, other trace species, aerosol particles, and meteorological parameters.

A working group was formed as a result of the workshop, consisting of experts, mostly from the USA, in monsoon circulations and transport, water vapour, and stratosphere/troposphere exchange. Additional participation by the international community is encouraged, particularly by scientists from south or south-east Asia directly affected by the summer monsoon, or those aware of local observations that would be of value to us.

The NH summer monsoons are uniquely interesting in the UT/LS for three reasons. (1) Much of the deep moist convection occurs over land, implying that cloud microphysical properties and the dynamics of convection itself differ significantly from tropical maritime convection. Evidence suggests that the role of convection in the water budget of the lower stratosphere differs between NH summer and winter, causing hydration of the layer in the summer, and dehydration in the winter. (2) Temperature soundings in the monsoon regions of NH summer have a tropical character, with a steep lapse rate and high tropopause near 100 hPa. These profiles and cloud observations imply that convective penetration of the UT/LS occurs equatorward of the jet stream. (3) Monsoon systems in NH summer are displaced relatively far from the equator, generate circulations asymmetric about the equator, with inter-hemispheric transport, and penetrate the lower stratosphere with meridional wind anomalies measurable up to the 30 hPa level. Upward penetration is more striking in the Asian summer monsoon than in the North American monsoon. Throughout the summer the jet stream is rather weak and constitutes an ineffective barrier to horizontal transport.

Satellite observations demonstrate a moistening of the lowermost stratosphere within or near the NH summer monsoon regions. There is an apparent connection between monsoon-induced water vapour anomalies and subsequent horizontal transport into the NH extratropics (on the west side of monsoon anticyclones) and tropics (on the east side of monsoon anticyclones). The tropical transport path is interesting insofar as the tropical lower stratosphere is a gateway to the entire middle atmosphere via the Brewer-Dobson circulation.

While certain details of these water-vapour transport paths may be accessible from existing observations, a special experiment to acquire *in situ* measurements in the UT/LS during NH summer has also been discussed. Such an experiment would complement recent and proposed studies of the cold trap during NH winter. Some insights might also be obtained on the so-called 'tape recorder' in stratospheric water vapour, and on whether summer monsoons have played a role in the observed trend of water vapour in the middle atmosphere, a trend unexplained by tropopause temperature.

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Announcement

SPARC Session at the 34th COSPAR Scientific Assembly

A session devoted to SPARC issues will take place at the 34th COSPAR Scientific Assembly, to be held 10-19 October 2002, in Houston Texas, USA. The session is:

• A1.1/C2.7 Climate Change Processes in the Stratosphere and at the Tropopause.

This session will focus on measurements, data analysis, modelling and theory related to processes near the tropopause and in the stratosphere that impact the global climate system. Particular attention will be paid to:

- 1) results or expected results from instruments that have been recently launched or that will be launched in the reasonably near future;
- 2) results and capabilities of new analysis methods, including data assimilation;
- 3) models indicating the roles of stratospheric processes in climate, with their implications for space measurements. Studies of transports across the tropopause, and other internal atmospheric barriers are also solicited.

The original of the abstract should be sent to the Copernicus Gesellschaft, with a copy to the Main Scientific Organizer, Dr.J.C. Gille.

Addresses and further information can be found at <http://www.copernicus.org/COSPAR/COSPAR.html>.

The Global Atmosphere Watch Monitoring Components

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Introduction

Monitoring of trace atmospheric constituents was originally driven by scientific curiosity. It was not long, however, before questions were raised as to what the consequences would be for humanity should the observed increases in certain trace chemicals continue unabated. The development of major international activities concerned with protection of the environment started in earnest during 1968 when the United Nations was called upon to organise a world conference on internationally significant problems related to the human environment. The conference took place in Stockholm in 1972 and successfully drew world-wide attention to environmental problems, including changes in the atmosphere due to rapid industrialisation.

During the 1970s three important atmospheric issues were addressed: (a) the threat of CFC's to the ozone layer, (b) acidification of lakes and forests in large parts of North America and Europe, caused principally by the conversion of sulphur dioxide into sulphuric acid by precipitation processes in the atmosphere and, (c) potential global warming caused by the build-up of greenhouse gases in the atmosphere. Each of these issues is now the subject of international treaties or conventions. The initial development of these agreements and the subsequent assessments of the mitigation measures they contain, rely heavily on the information derived from WMO's atmospheric composition monitoring programme.

The WMO formally embarked on a programme of atmospheric chemistry and meteorological aspects of air pollution during the 1950s. This included assuming responsibility for standard procedures for uniform ozone observations and establishing the Global Ozone Observing System (GO₃OS) during the 1957 International Geophysical Year. In the late 1960s, the Background Air Pollution Monitoring Network was established which was subsequently consolidated with the Global Ozone Observing System into the current WMO Global Atmosphere Watch (GAW) in 1989. The GAW monitoring programme

includes a co-ordinated global network of observing stations along with supporting facilities. GAW provides data for scientific assessments and for early warnings of changes in the chemical composition and related physical characteristics of the atmosphere that may have adverse affects upon our environment. Monitoring priorities have been given to greenhouse gases for possible climate change, ozone and ultraviolet radiation for both climate and biological concerns, and certain reactive gases and the chemistry of precipitation for a multitude of roles in pollution chemistry.

The GAW programme was briefly described in SPARC Newsletter N° 7 (1996) with emphasis on the very successful GO₃OS. The GAW programme has matured considerably since then, and presently consists of many co-ordinated components that have been designed to provide accessible, high quality atmospheric data to the scientific community. These components include: (a) measurement stations, (b) calibration and data quality centres, (c) data centres and (d) external scientific groups for programme guidance. **Figure 1** shows the principal components making up the GAW programme. Support for these components is provided, in large part, by individual WMO Member countries that directly partici-

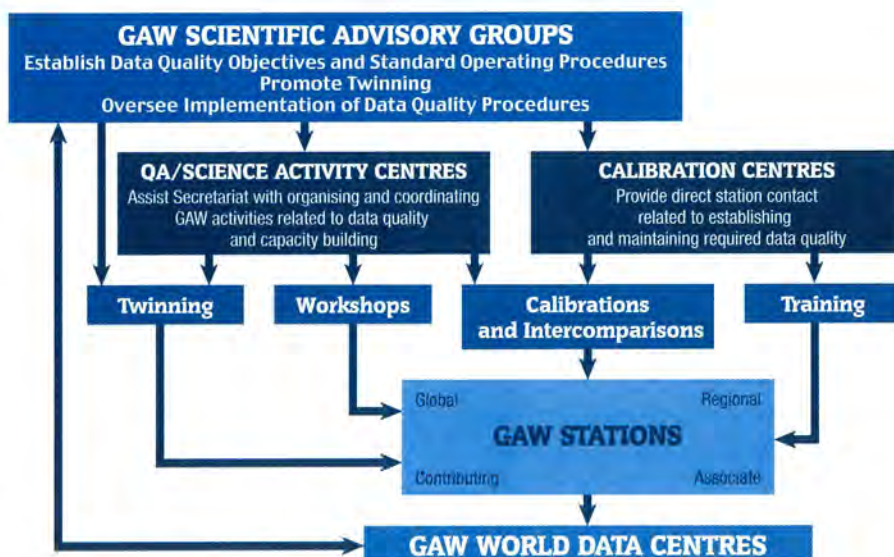
pate in the GAW programme, augmented by some outside international funding, and the WMO Secretariat's internal budget.

Measurement stations

A network of measurement stations is the backbone of the GAW programme. This network consists of GAW Global and Regional measurement stations with additional measurements from Contributing and Associate stations. The present network of 22 GAW Global stations is shown in **Figure 2** (p. III). Global stations are usually situated in remote locations, representative of large geographic areas, have very low (background) levels of pollutants, and continuously measure a broad range of atmospheric parameters over decades. It is important to note that Global station sites must be entirely free of the effects of local and regional pollution sources for substantial periods throughout the year. Data are typically applied to global issues such as climate change and stratospheric ozone depletion.

GAW Regional stations are usually representative of smaller geographic regions. Regional station sites are often chosen so they are not affected by nearby sources of pollution such as vehicles, industrial combustion or agricultural

Figure 1: Monitoring components making up the GAW programme.



activities. Measured parameters vary considerably at Regional stations, and depend upon the regional needs. Some Regional stations measure a broad range of parameters, while others measure only a single measurement type. Not all atmospheric parameters can or even should be measured at each GAW Station. For example, a site with excessive local pollution may still be suitable for column ozone and solar radiation but not for background measurements of greenhouse gases. Data from Regional stations are typically applied to more local or regional issues such as acid deposition, transport of trace gases and aerosols, regional background pollution levels, and local UV or ozone conditions. Both Global and Regional stations are operated by their host countries, either by their National Meteorological Services or by other national scientific organisations. More than 65 countries actively host GAW stations.

In addition, there are stations that have not been formally designated as either Global or Regional GAW stations, but nevertheless make very valuable contributions to the GAW programme through submitting data to the GAW World Data Centres (WDCs). Such stations are classified as either Contributing stations or Associate stations. Contributing stations belong to other organisations such as the Network for Detection of Stratospheric Change (NDSC), the European Monitoring and Evaluation Programme (EMEP), or the Baseline Surface Radiation Network (BSRN). Through mutual agreements, these contributing organisations are under the GAW umbrella of stations. Some of these Contributing stations have dual affiliations within GAW, having also an established Global or Regional GAW station status. And finally, there are the Associate stations that submit data to the WDCs without any formal GAW affiliation. These include many single measurement stations as well as air and ship platforms.

Calibration and data quality centres

The calibration and the data quality centres are either regional or global in responsibility. These centres perform the vital function of helping to ensure that the data submitted to the GAW World Data Centres is of sufficient quality to meet the needs of the scientific research community. Their activities include (a) careful calibration of instruments through station visits,

(b) instrument intercomparisons and calibration campaigns, (c) laboratory measurement intercomparisons of circulated standard gases or reference samples, and (d) systematic and frequent calibration checks of the world standards. Whenever possible, measurement calibrations are traceable to the GAW world calibration standards.

Although these direct calibration activities are required to maintain the required comparability of measurements globally, the GAW data quality programme includes other activities. For example, during station visits, GAW Calibration Centres provide on-site training and expert advice to help station personnel maintain the required data quality. Workshops are also held during intercomparisons and calibration campaigns, while instrument repairs or upgrades are often performed on the spot by technical experts. Further help is provided to the less experienced personnel at new stations by personnel from well established and technically advanced stations. In addition, there are GAW training centres sponsoring frequent training sessions for station personnel, particularly those located in developing countries.

World Data Centres

The GAW World Data Centres (WDCs) are operated and maintained by their individual host institutions. They collect, document and archive atmospheric measurements and the associated metadata from measurement stations world-wide and make these data freely available to the scientific community. In some cases, WDCs also provide additional products including data analyses, maps of data distributions, and data summaries.

Presently, there are six GAW WDCs each responsible for archiving one or more GAW measurement parameters or measurement types. Measurement types presently archived include ozone (surface, total column, vertical column), greenhouse and carbon cycle gases, oxides of nitrogen, chemistry of precipitation, chemical and physical properties of aerosols, solar radiation including ultraviolet, and station meteorological parameters. The data residing in the WDCs are always subject to change, for example, from new calibrations. For this reason, data providers at individual stations should always be contacted directly for detailed information on such changes. In order that data providers be given appropriate recognition, scientists

accessing the data are required to accept the following conditions: "For scientific purposes, access to these data is unlimited and provided without charge. By their use you accept that an offer of co-authorship will be made through personal contact with the data providers or owners whenever substantial use is made of their data. In all cases, an acknowledgement must be made to the data providers or owners and the data centre when these data are used within a publication."

The World Ozone and Ultraviolet Radiation Data Centre (Toronto, Canada)

The World Ozone and Ultraviolet Radiation Data Centre is operated by the Experimental Studies Division of the Meteorological Service of Canada (MSC), until recently known as the Atmospheric Environment Service, Environment Canada. The Data Centre began as the World Ozone Data Centre (WODC) in 1960 and in June 1992, the MSC agreed to a request from the WMO to add ultraviolet radiation data to the WODC. The Data Centre has since been renamed the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) with the two component parts: the WODC and the World Ultraviolet Radiation Data Centre (WUDC).

The WODC operates a scientific archive and database providing a variety of ozone data sets to the international scientific community. Some data records date back more than 40 years. Data sets include total column ozone and vertical profile data from ozone-sonde flights, lidar measurements and the Umkehr technique. Archived data are available by conventional FTP access or by a direct link from the WOUDC Data Web page. At present, access to the archive by conventional FTP requires a username and password, which is available upon request. Value added data products include total ozone time series graphs and near real-time ozone maps.

The WUDC like its ozone counterpart, is a scientific archive and database of high resolution spectral, multi-band and broad-band UV data. The database started receiving data in early 1993.

At present, there are over 400 registered stations represented in the WOUDC archive. Metadata information such as station lists with locations, are available from the WOUDC Web site: <http://www.msc.ec.gc.ca/woudc>.

The World Radiation Data Centre (St Petersburg, Russian Federation)

The World Radiation Data Centre (WRDC) is located in St. Petersburg at the Main Geophysical Observatory of the Russian Federal Service for Hydrometeorology and Environmental Monitoring. The WRDC was established in 1964, and since that time it centrally collects, archives and publishes radiometric data for the world, to ensure the availability of these data for research purposes by the international scientific community. It issues a periodic bulletin entitled "Solar Radiation and Radiation Balance Data. The World Network."

The WRDC processes solar radiation data currently submitted from more than 500 stations located in 56 countries and operates an archive with more than 1200 stations listed in its catalogue. The WRDC is the central depository of the measured components such as: global, diffuse and direct solar radiation, downward atmospheric radiation, net total and terrestrial surface radiation (upward), spectral radiation components (instantaneous fluxes), and sunshine duration, on hourly, daily or monthly basis.

At the present time, recent and current data can be viewed by accessing the WRDC web site. It also contains a list of all stations that have submitted data. As new measurement data are received from National Meteorological Services and other institutions in paper form, on magnetic media or electronically, the WRDC performs a supplementary quality control and requests stations to confirm the data for total quality assurance. The data from GAW sites is represented in a specified directory as tables and as figures including graphs showing relationships between measured and estimated parameters: (<http://wrdc.mgo.rssi.ru>).

The World Data Centre for Precipitation Chemistry (Albany, USA)

The responsibility for the World Data Centre for Precipitation Chemistry (WDCPC) was originally given to the US Environmental Protection Agency (EPA) which operated the programme until 1994. The responsibility was subsequently transferred to the Atmospheric Sciences Research Center (ASRC) of the State University of New York at Albany, and is supported by US government agencies.

The Data Centre is responsible for documenting, archiving and making available data on the chemical components of precipitation that has been collected at hundreds of stations around the world. The precise chemical composition of each sample is determined by collaborating laboratories working within strict guidelines and standards developed under GAW.

The samples are analysed for many chemical constituents and its acidity or alkalinity. Specifically, the chemical constituents of interest are: sodium, potassium, magnesium, calcium, fluoride, chloride, bromide, ammonium, nitrate, sulphate, sulphur, phosphate tribasic, mercury, aluminium, cadmium, copper, iron, manganese, nickel, lead, zinc, phosphorus, iodine, nitrite, and sulphite (http://marble.asrc.cesim.albany.edu/qasac/gaw_maps.html).

The World Data Centre for Greenhouse Gases (Tokyo, Japan)

The World Data Centre for Greenhouse Gases (WDCGG) was established at the Japan Meteorological Agency (JMA) in October 1990 and collects and distributes data on the mixing ratios of greenhouse (CO_2 , CH_4 , CFCs, N_2O , etc.) and related reactive (CO , NO_x , SO_2 , VOC, etc.) gases in the atmosphere and the ocean. Simultaneously measured meteorological parameters are also recorded at WDCGG.

Data contributors submit their data on diskettes, coding sheet, or via FTP according to a prescribed format. As of February 2000, 182 stations in 42 countries submitted observational data for 13 species of greenhouse and related gases to the WDCGG. The WDCGG periodically publishes the "WMO WDCGG Data Report", "WMO WDCGG CD-ROM", "WMO WDCGG Data Catalogue" and "WMO WDCGG Data Summary". The "Data Report" contains monthly and annual means and the "CD-ROM" also contains hourly and daily means. Information on contact persons, methods of measurement and calibration at the observation stations is also made available, and all data can be accessed by FTP file transfers. The "Data Catalogue" provides an index of existing data along with procedures for data access. It also includes information on site environment, measurement procedures, and calibration techniques. The "Data Summary" includes averaged results from the past and present conditions on local, regional, and global scales (<http://gaw.kishou.go.jp/wdcgg.html>).

The World Data Centre for Aerosols (Ispra, Italy)

The World Data Centre for Aerosols (WDCA) is operated by the European Union's Joint Research Centre, Ispra, Italy, and was set up to archive aerosol related observations made under GAW. A wide variety of aerosol parameters have been recommended by the GAW Scientific Advisory Group for Aerosols as suitable for measurement and archiving at GAW Regional and Global stations.

Recommended aerosol parameters for frequent measurement at Regional stations:

- Multiwavelength aerosol optical depth.
- Mass, preferably in two-size fractions (either 10-micron ambient and 1-micron dry, or 10-micron ambient and 2.5-micron ambient).
- Major chemical components in the above two size fractions.
- Light scattering coefficient (Nephelometry).

Additional aerosol parameters recommended for frequent measurement at Global stations:

- Scattering and hemispheric backscattering at various (3 or 4) wavelengths.
- Aerosol number concentration.
- Cloud condensation nuclei at 0.5% supersaturation.
- Diffuse, global and direct solar radiation. (These measurements are archived at WRDC)

Parameters for intermittent measurement:

- Aerosol size distribution.
- Detailed size fractionated chemical composition.
- Size dependence on relative humidity.
- CCN spectra at various supersaturations.
- Vertical distributions of aerosol properties.

Clearly, few stations can make all of these measurements, while many stations are making only one or two of these observations. At the moment, WDCA is concentrating on building up the archives of commonly available parameters, including chemical composition, aerosol optical properties, and CN concentrations (<http://www.ei.jrc.it/wdca/>).

The World Data Centre for Surface Ozone (Kjeller, Norway)

The World Data Centre for Surface Ozone (WDCSO) was formally established in June 1997 and is the newest of the six WDCs. The Data Centre has

been operational since August 1998 as an integral part of the Norwegian Institute for Air Research's (NILU) Atmospheric Database for Interactive Retrieval (NADIR). WDCSO provides a quality archive for surface ozone measurements originating from the WMO-GAW umbrella of monitoring stations and from other measurement programmes and campaigns (<http://www.nilu.no/projects/nadir/wdco3/wdco3.html>).

External scientific groups

The WMO Commission for Atmospheric Sciences (CAS) is the lead WMO technical commission for research activities (including those of GAW) in the atmospheric sciences and related fields. The Executive Council Panel of Experts/CAS Working Group on Environmental Pollution and Atmospheric Chemistry (EC Panel) has been designated as the overall steering body for GAW. The EC Panel is composed of internationally known experts that are representative of a broad range of interests in atmospheric chemistry relevant to the GAW programme.

The EC Panel has established Scientific Advisory Groups (SAGs) to organise and co-ordinate GAW activities for five parameter types: ozone, greenhouse gases, UV, precipitation chemistry, and aerosols. Each SAG is responsible for establishing and overseeing the implementation of data quality criteria and standard operating procedures for its assigned parameter type. In addition, the SAGs are to promote capacity building through training and education at GAW stations in developing countries, particularly during the early operation at newly established stations. This is done through matching those stations or individuals in need of help with experienced laboratories or scientists to provide the needed support. Such "twinning" of the experts with the inexperienced is a critical component to achieve high quality data from stations in countries without the local expertise.

Concluding perspectives

GAW is implemented in accordance with the principle that when a country agrees to participate, full responsibility for its GAW activities rests with that country. WMO does not, for example, cover the costs of long-term maintenance of GAW stations and facilities. Nevertheless, many developing countries require outside support to start, or improve their participation in the GAW

programme. In recognition of this, WMO has provided limited support for developing countries through a variety of methods whenever funds are available. Capacity building is of critical importance to GAW and is provided through workshops, reports, consultant services, and onsite training by outside experts. WMO also has encouraged its Member countries to include GAW projects in their UNDP national programmes, and to enter "twinning" arrangements where, on a bilateral basis, a laboratory, institute or an experienced individual scientist sponsors measurements of specific parameters at a station or group of stations in developing countries. Whenever funds can be made available, the WMO also provides limited support for the maintenance of instruments through calibrations, repairs and upgrades. Also, measurements have been initiated in data sparse regions and new GAW stations have been established by the WMO through international funding mechanisms.

Space-based observations offer a partial solution toward the goal of achieving global coverage of certain measurements important to the GAW programme. These measurements include ozone (column and profile), solar radiation, and surface measurements of some trace gases such as carbon monoxide as well as ancillary parameters. Satellite observations are of benefit to the global community and are of special interest to those geographic regions where there are gaps in GAW's surface-based monitoring network. For this reason integration of satellite observations into the GAW programme is highly desirable. In turn, satellite systems can best meet their established requirements if they are checked against highly accurate ground based measurements of known quality. The GAW network of stations have provided both total column ozone and vertical profile ozone data for satellite validation for the past few decades. A new generation of satellites sensors have begun operation and presently include the capability to make free tropospheric measurements of carbon monoxide and column methane. A few of the high altitude GAW stations measure carbon monoxide and could be of value for checking the accuracy of these relatively new satellite measurements.

In addition to the GAW monitoring programme, the WMO GAW Urban Research Meteorology and Environment (GURME) project was recently started in response to the requests of the National

Meteorological and Hydrological Services (NMHSs). The lead responsibility for the programme rests with the Scientific Advisory Group (SAG GURME). NMHSs have an important role to play in the study and management of urban environments because they collect information and have capabilities that are essential to the forecasting of urban air pollution and the evaluation of the effects of different emission control strategies. The WMO established GURME as a means to help enhance the capabilities of NMHSs to handle meteorological and related aspects of urban pollution. GURME is designed to do this through co-ordination and focussing of present activities, as well as initiation of new ones.

In April 2001, the EC Panel approved a new strategic plan for GAW and is given in the WMO/GAW Report #142 "Strategy for the Implementation of the Global Atmosphere Watch Programme (2001-2007)". This report includes a list of 8 strategic goals:

- (a) Improve measurements programme for better geographical and temporal coverage and for near real time monitoring capability,
- (b) Complete the quality assurance/quality control system,
- (c) Improve availability of data and promote their use,
- (d) Improve communication and co-operation between all GAW components and with the scientific community,
- (e) Identify and clarify changing roles of GAW components,
- (f) Maintain present and solicit new support and collaborations for the GAW programme,
- (g) Intensify capacity building in developing countries,
- (h) Enhance the capabilities of NMHSs in providing urban-environmental air quality services.

Over the lifetime of this Strategic Plan, human activities will continue to change the composition of the atmosphere, although efforts will be made to control them. The WMO GAW network of stations must continue to monitor the critical atmospheric parameters and provide scientific data needed to understand and ultimately predict environmental changes on both regional and global scales. As GAW continues to mature, closer co-operation with SPARC and with other organisations closely tied to the use of these data, needs to be developed.



European Research in the Stratosphere, 1996-2000: Advances in our Understanding of the Ozone Layer during THESEO

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An assessment of European research on stratospheric ozone and UV radiation [1] has recently been published by the European Commission (EC) (see **Figure 1**, p. IV). It contains a thorough review of the progress of the European research programme on stratospheric ozone and UV radiation between 1996 and 2000 and assesses the contribution of European research to the current state of knowledge. It thus highlights the major achievements of the co-ordinated EC and nationally funded research programmes. The results from the Third European Stratospheric Experiment on Ozone (THESEO), a major part of European stratospheric research in this period, feature strongly. It is hoped that this document will be a valuable resource for scientists working on the stratosphere and particularly for the preparation of the 2002 WMO-UNEP Ozone Assessment. It has taken over 100 scientists almost two years to prepare this assessment and it was subject to peer review by an international group of scientists. A similar assessment for the period 1989-95 was published in 1997 [2].

The structure of the report is 'issue oriented' so that the main scientific advance are presented with reference to the overall stratospheric ozone problem. The chapter titles and lead authors are:

1. Introduction:
G. Megie and G.T. Amanatidis
2. Long term changes:
M. Van Roozendaal and G. Vaughan
3. Polar ozone:
K.S. Carslaw and H. Oelhaf
4. Mid-latitude and tropical ozone:
A. Hauchecorne and Th. Peter
5. Stratospheric ozone and the link to climate change:
J. Austin and U. Langematz
6. Atmospheric UV radiation:
M. Blumthaler and P. Taalas
7. Aviation impact on atmospheric composition and climate:
U. Schumann and J. Ström

The main findings are contained in an executive summary and these in turn are summarised:

- Large losses (20-30% in the ozone column, 40-70% at altitudes around 18 km) have occurred in the Arctic during each of the five cold stratospheric winters since 1993/94. Recent campaigns, including THESEO, have revealed a great deal about the role of halogen compounds (from CFCs and other ozone-depleting gases), particles and stratospheric meteorological conditions that lead to the large ozone losses in the Arctic and the subsequent reduced ozone amounts over Europe and the northern mid-latitudes.

- Stratospheric ozone amounts over mainland Europe started to decline in the 1970s, with the biggest decrease (5-10% in the ozone column) occurring in winter and spring. Biologically active UV has increased at the ground in line with the reduced ozone amounts. The ozone decline has resulted from (a) chemical processes resulting from the break-down of CFCs and other ozone depleting gases and (b) decadal changes in stratospheric meteorology, which are much better understood. For example, the frequency of ozone 'mini-holes' over Europe has increased and is linked to climatic changes in atmospheric circulation in the north Atlantic and European region.

- The existing international controls on ozone-depleting gases agreed through the Montreal Protocol process have caused chlorine amounts to start to decrease in the atmosphere. However, the growth in bromine concentrations continues. Continued increase of halons over the next few years could cause the abundance of equivalent chlorine to decline more slowly than predicted.

- Despite the current levelling off in the concentration of ozone depleting substances limited under the Montreal Protocol, any ozone recovery in the northern hemisphere is likely to become measurable around 2010, at the earliest, due to the large atmospheric variability. Full ozone recovery is expected when the chlorine amounts

in the atmosphere reach the pre-ozone hole levels in 50 or so years.

- However, future stratospheric ozone and UV levels will depend not only on the successful control of chlorine and bromine concentrations, but also on the impact of climate change and other trace gases such as N₂O, H₂O and CH₄ on the stratosphere. A continuation of the observed cooling of the stratosphere may extend the period in which severe wintertime depletion of ozone over the Arctic can occur. Changes in climate are also likely to affect the stratospheric ozone distribution over mid-latitudes.

- The atmospheric impact of the present fleet of aircraft has been thoroughly studied. The role of the nitrogen oxides in producing ozone is much better quantified and there has been a great deal of progress in the evaluation of the importance of the climate effects of aircraft contrails. Achieving a similar understanding of the aviation influence on cirrus clouds remains a major challenge.

Anyone interested in obtaining a copy should contact G.T. Amanatidis, EC DG Research, Rue de la Loi 200, B-1049 Brussels, Belgium; fax: 32 2 296 3024; email: georgios.amanatidis@cec.eu.int.

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- [1] European Research in the Stratosphere, 1996-2000: Advances in our understanding of the ozone layer during THESEO, eds. G.T. Amanatidis and N.R.P. Harris, European Commission, EUR 19867, 2001.
- [2] European Research in the Stratosphere - The contribution of EASOE and SESAME to our current understanding of the ozone layer, European Commission, EUR 16986, 1997.

Report on the Second IAGA-ICMA-PSMOS Workshop on Long Term Changes and Trends in the Atmosphere

Prague, 2-6 July 2001

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Programme Committee: G. Beig (India), A.D. Danilov (Russia), M.-L. Chanin (France), J. Lastovicka (Czech Republic, chairman), J. Stegman (Sweden), G. Thomas (USA, vice-chairman).

The Workshop was organised by the Institute of Atmospheric Physics (IAP) of the Academy of Sciences of the Czech Republic and co-organised by the Department of Meteorology and Environmental Protection of Charles University under auspices of the IAGA/ICMA/PSMOS working group "Long Term Changes and Trends in the Mesosphere, Thermosphere and Ionosphere" (LT-TIME). About 50 scientists from 17 countries presented 47 oral papers, including 19 invited and invited review papers. Refereed papers will be published in a special issue of the journal *Physics and Chemistry of the Earth*.

The Workshop has been supported by IUGG, SPARC, a Czech company (Pražská plynařenská), and IAP. EGS, IAGA, ICMA/IAMAS and SCOSTEP have provided moral and informational co-sponsorship.

The focus was on trends in the mesosphere and ionosphere, but trends in the thermosphere, stratosphere and troposphere were treated as well.

Selected results

The Earth's orbital variation will be extremely small in the next 50 000 years, which is expected to result in an unusually long interglacial period. The doubling of the CO₂, with respect to present concentration would result in the disappearance of the Greenland ice sheet in about 4 000 years.

The global water vapour concentration in the stratosphere is reported to increase by about 0.05 ppmv/year. Satellite observations confirm cooling of the whole stratosphere, which is significant above about 3 hPa but rather marginal below. Increasing CO₂, decreasing stratospheric ozone and increasing water vapour contribute to this cooling. The effect of the solar cycle in stratospheric temperatures is important, therefore data sets for trend investigations should be at least 20 years long.

Arguments were presented indicating that ozone trends in the lowermost stratosphere observed at Payern (Switzerland) are predominantly of dynamical origin. Also the longitudinal variation of trends at mid-latitudes in the Atlantic region seems to be mainly of dynamical origin. About a quarter of the ozone decrease at mid-latitudes in Europe could be caused by a long-term change of the AO, almost half of this decrease is related to changes of height of the 100 hPa level (the influence of these two factors probably overlaps). The overall ozone content in laminae in ozone profiles at 36-75°N had decreased from the late 1960 to early 1990 by 50%. In combination with a strong seasonal variation of number of laminae, this contributed substantially to the seasonal variation of trends in total ozone.

The round-table discussion on trends in mesospheric temperature made it clear that the differences between the results obtained by different authors may be due to the dependence of the trends on latitude and longitude, altitude, season and time of the day. The differences in the length and date of analysed periods also play some role, as do instrumental and other technical problems.

The first draft version of the executive summary on the published papers dealing with trends in mesospheric and mesopause region temperatures has been presented by the MTTA (Mesospheric Temperature Trend Assessment) panel constituted under the auspices of the working group which organised this workshop. In the mid-latitudes the lower mesosphere shows a trend of around -2°K per decade in several studies, but in the middle mesosphere rockets results reveal a larger cooling (-7°K/decade) than lidar (-3°K/decade). Contradictory results are reported for the mesopause region. The trend in the summer mesopause is small or close to zero. Rocket launches in Scandinavia in summer do not

reveal any trend of temperature at 82 km (the noctilucent cloud height). Model computations provide a twice as large response of mesospheric temperature to the increasing greenhouse gas content in winter as compared to summer. Airglow emission measurements in Sweden provide even larger winter-summer difference in temperature trends. The implied shrinking of the mesosphere deduced from temperatures inferred from airglow measurements is about 50 m/year near 70 km (increased to 120 m/year near 100 km).

The water vapour content in the mesosphere was increasing since the 1950s by about 1%/year. Such an increase cannot be fully explained by the increasing concentration of methane. HALOE measurements at 75 km over 1991-2001 revealed a positive trend in ozone concentration of 2-3 %/decade at sunrise but no detectable trend at sunset. From measurements at Collm, the prevailing wind rather strengthened over 1980-2000, which contradicts the results from some other stations.

Our knowledge and understanding of trends in the lower ionosphere is insufficient. The available results indicate an increase in electron density at lower altitudes as a consequence of changes in chemistry of minor constituents and the thermal shrinking of the mesosphere due to greenhouse cooling.

Trends in the E and F1 layers of the ionosphere seem to be reasonably consistent with model expectations: foE and foF1 increases, h'E decreases. Results of different authors on the F2-layer ionosphere trends differ substantially even as to sign.

Reports on the IAMAS General Assembly

10-18 July 2001, Innsbruck, Austria

The Middle Atmosphere Chemistry, Transport, and Radiation symposium

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A six-day symposium on Middle Atmosphere Chemistry and Transport was organised under the banner of ICMA. The conveners were: A. R. Ravishankara (Boulder, USA), Martyn Chipperfield (Leeds, UK), Victor Fomichev (Toronto, Canada) and Darryn Waugh (Baltimore, USA). It was organised to cover the following themes:

1. Ozone trends in the stratosphere and the upper troposphere
2. Stratospheric and tropospheric processes
3. Stratospheric-Troposphere interactions
4. Mesosphere/upper stratosphere processes (2 sessions)
5. Stratospheric polar processes (2 sessions)
6. Measurements of atmospheric constituents
7. Laboratory studies of atmospheric processes

Each of the nine sessions, had two invited speakers. A total of 18 invited talks and 40 contributed talks were presented. Overall the discussions following the talks were lively. In addition, a poster session was also included and it had 11 posters. To give a flavour of the meeting, we have included brief summaries of the invited talks and some of the contributed talks.

Ozone trends

The symposium started with a session on trends in ozone in the stratosphere and the upper troposphere (UT). Currently, this is a topic of major interest. **R. Hudson** presented an analysis of satellite derived stratospheric column ozone abundance from 1979 to 1992. He had averaged the data according to different 'dynamical' regimes from the

mid-high latitudes based on meteorological features of the troposphere and lower stratosphere (LS). Interestingly, when analysed in these regimes, the observed downward trend in mid-latitude ozone mostly disappeared, suggesting, rather contentiously, that these trends are not related to chemical loss but to the movement of dynamical regimes. **J. Logan** presented an analysis of ozone sonde data for the UT/LS for stations over Europe and Canada. The observations near 18 km showed a clear downward trend. However, in the lowermost stratosphere, a difficult region to analyse, the variability was much larger with less indication of an overall trend. These ozone trends and their analyses will be a major component of the upcoming WMO/UNEP ozone assessment.

Other presentations discussed the effects of dynamics and chemistry on the trends in column ozone and the effects of ozone changes on the stratosphere.

Mesospheric and upper stratospheric processes

M. Mlynczak presented a detailed estimate of how well we can observe the atmospheric energy budget from the edge of space down to the surface. He stated that with a forthcoming TIMED satellite observations we will be able for the first time to directly observe or derive from appropriate measurements the rates of solar heating and infrared cooling throughout the entire atmosphere below 100 km. This was augmented by contributed talks on the energy budget produced by the extended Canadian Middle Atmosphere Model from the tropopause up to the lower thermosphere and latitude-altitude distribution of the infrared cooling calculated with use of temperature, atomic oxygen and CO₂ data retrieved from the CRISTA limb radiance measurements.

V. Ogibalov presented an overview of the current state of the non-LTE problem for the vibrational rotational CO₂ bands and discussed features and accuracy of recently developed non-LTE models.

Papers presented by **M. Kaufmann** and **S. Melo** dealt with measurements of temperature structure and composition

of the mesosphere and lower thermosphere region. Kaufmann presented results on temperature, O₃ and CO₂ obtained during two CRISTA missions. The CO₂ mixing ratio in the 70-100 km region is significantly smaller than that shown by previous rocket borne mass spectrometer data and shows considerable latitudinal variation. Temperature and CO₂ profiles retrieved from the first CRISTA mission were also presented during contributed talks and shown to be in good agreement with those presented by Kaufmann. In addition, a new algorithm for the retrieval of kinetic temperature from measurement of CO₂ limb emission was also presented.

Other talks addressed the role of precipitating electrons and solar proton events in affecting O₃ abundance in the upper stratosphere and mesosphere and the impact of time dependent O₃ and Sea Surface Temperature in numerical simulation of the middle atmosphere.

Stratosphere/Troposphere interactions

T. Peter presented lidar measurements made during the APE-THESEO aircraft campaign based out of Mahe/Seychelles (5S) in February/March 1999. They show ultra-thin subvisible clouds near the tropical tropopause. It was argued that large upwelling (around 5mm/s) covering a broad region is required to explain the existence and stability of these clouds. Some evidence for large upwelling in ECMWF analyses was also presented. **C. M. Volk** also presenting measurements from the APE-THESEO campaign, focused on *in situ* tracer measurements (N₂O, CFCs, SF₆, and O₃) made just above and below the tropopause. The vertical profiles of the tracers show almost constant values in the UT (340-380K), indicating little mixing of stratospheric air into this region. Assuming no mixing and specified production rates, upwelling of around 2mm/s was inferred from the measured ozone profiles. Latitudinal gradients in the tracers were also examined, and tracers with large inter-hemispheric surface gradients (e. g., SF₆) were shown to still have significant gradients in the upper troposphere.

Other presentations discussed the observations of water vapour in the tropical UT, tracer correlations near high latitude UT, and models of chemistry-climate feedbacks. There were also talks on cross tropopause exchanges of species and the impact of solar cycle on stratospheric circulation.

Measurements of atmospheric constituents

The measurements of atmospheric species, a key part of atmospheric research, included invited talks by **J. Burrows** and **P. Wennberg**. Burrows demonstrated the capacity of satellite-based instrument for providing much needed data on a global scale for many stable and reactive species. He exemplified the capabilities by using the data from the GOME satellite (**Figure 1**, p. IV). It included data on stable species such ozone and nitrogen dioxide as well free radicals such as BrO. The ability of the satellite to get information on tropospheric composition was a significant showing. Wennberg concentrated on local measurements of free radicals such as OH and HO₂, in conjunction with many other stable species from the ER-2 aircraft. He demonstrated the power of such measurements in getting key information on atmospheric processes, including the ability to constrain the uncertainties in reaction rate coefficients. He highlighted his talk by showing the importance to overtone photodissocia-

tion of HO₂NO₂ as source of odd-hydrogen in the lower stratosphere (**Figure 2**).

Satellite derived evidence for meridional mixing of minor constituents, use of DOAS in satellite measurements, algorithms for aerosol retrieval, and *in situ* measurement of chlorine oxides in a rocket plume were also presented.

Stratospheric and tropospheric processes

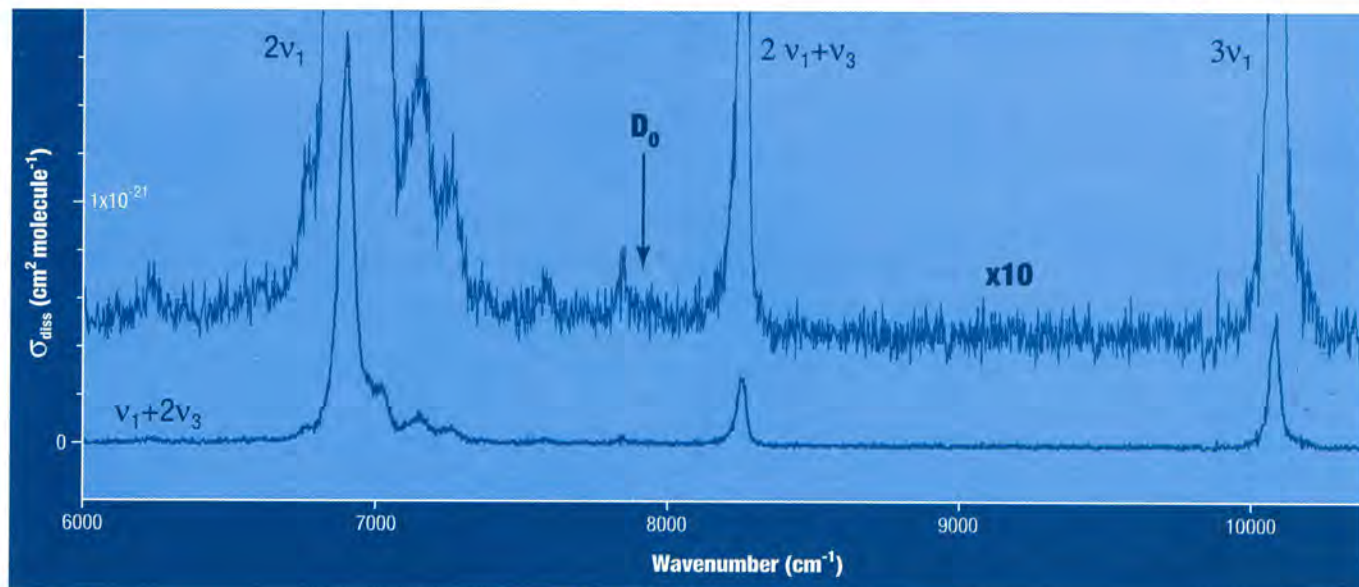
The heart of middle atmospheric chemistry is the processes that control the ozone level and the radiation balance in this region. Therefore, two invited talks and many contributed talks addressed these issues. **S. Lal** presented vertical distributions of several trace gases (e. g., N₂O, CH₄, CFCs, and SF₆) from balloon flights from Hyderabad, India (17. 5°N, 78. 6°E) made between 1985 and 1998. Several important quantities were calculated, including the age of the air (from SF₆), the total amount of organic and inorganic chlorine, and the lifetime of several of the tracers (using known lifetimes of other tracer and tracer-tracer correlations). **D. Hartmann** presented calculations from a radiative transfer model that show that thin cirrus near the tropical troposphere are radiatively cooled if they lie above thick convective anvil clouds. Hartmann argued that this cooling leads to subsidence and low tropopause temperatures, and that quasi-horizontal advection through these cold

regions could be an efficient mechanism for dehydration of stratospheric air. Other talks addressed the recovery of chlorine as seen from satellite data, the variability of stratospheric sulphate abundance, and variation of nitrogen oxide concentrations. Some talks addressed the variability of the stratosphere as seen in GCM simulations, tracer-tracer correlation, age of air, distribution of BrO and its impact on ozone, and the simulation of Mt. Pinatubo aerosol.

Stratospheric polar processes

The current heightened interest in the Arctic ozone changes in the stratosphere led to a session with four invited talks and many contributed talks. First, **D. Fahey** presented measurements from the ER2 aircraft made during the SOLVE/THESEO campaign in 1999/2000, which show extensive denitrification, chlorine activation, and chemical ozone loss within the Arctic vortex. These measurements also indicate large HNO₃-containing particles ("rocks"), up to 20 micron in diameter (**Figure 3**), which provide an effective mechanism to denitrify the LS. Fahey also presented an analysis of ozone and NO₂ loss that showed an enhancement of chemical ozone loss in denitrified air parcels. **P. Newman** presented the results from an analysis of LS polar temperatures and eddy heat fluxes using NCEP/NCAR re-analyses. This analysis showed that the spring polar temperatures are controlled by the integrated wave activity over the

Figure 2. Wennberg showed that in the UT/LS the IR photodissociation cross sections of HO₂NO₂ (Figure below) yields a photolysis rate of $\sim 1 \times 10^{-5} \text{ s}^{-1}$, independent of the solar zenith angle. In much of the lower atmosphere, this process is often the dominant photochemical loss mechanism for HO₂NO₂. Photochemical models that ignore this process, overestimate the concentration of HO₂NO₂ and thereby overestimate the loss rate of OH via OH + HO₂NO₂. The figure shows the measured photodissociation cross sections for HO₂NO₂ in the near IR. Photodissociation in this spectral region is dominated by excitation of the overtone of the OH stretching frequency near 7000 cm⁻¹. The quantum yield for photodissociation is less than unity because the energy of these photons is less than the 0 K bond dissociation energy, D_0 .



preceding 45-60 days, and that the recent cold northern winters are a result of this weakened wave driving of the stratosphere. Output from several GCMs was also analysed, and model-data biases were noted. There were a total of 11 presentations (including 4 invited and 1 poster) dealing with processes in the upper stratosphere-lower mesosphere region. **M. Rex** presented results from the MATCH technique for diagnosing Arctic ozone loss for the past 10 years. The derived loss correlates with the availability of sunlight as expected. The loss also shows an extremely tight, almost linear, correlation with the area available for PSC formation - a tighter correlation than might be expected when considering other atmospheric variability. The last invited talk in this session was by **F. Goutail**. She also discussed Arctic O_3 loss based on comparisons of ground-based and satellite data with models. She showed that under certain conditions the models could reproduce the observed loss well, while in other winters the models have a tendency to underestimate the loss.

This session on the polar stratosphere also included contributed talks on dehydration signals, the origin of polar vortex air, *in situ* observations of ClO and its implied value for the rate coefficient for the ClO self reaction, simulation of ozone depletion using a Lagrangian model, ozone changes in a climate model along with its impact, and non-linearities in a gas phase model of the polar chemistry.

Laboratory studies of atmospheric processes

In the session dealing with laboratory studies, which form the underpinning of our understanding of the atmosphere, there were two invited talks; the first dealt with gas phase chemical kinetics and second with heterogeneous reactions. **P. Wine** updated the current understanding of the rate coefficients for reactions of nitrogen species in the stratosphere. It ranged from the reactions of $O(1D)$ with HCN and CH_3CN to the reaction of O atoms with NO_2 . The talk highlighted how certain key stratospheric reactions have been defined to very high accuracy such that the uncertainties associated with the rate coefficients of such reactions are negligible in defining stratospheric ozone depletion. **J. Abbatt** exemplified the basic steps involved in the heterogeneous reactions on ice. The talk showed how surface uptake/reactions may be enhanced two reactants that react together are simultaneously exposed to ice surface. The reaction that was highlighted involved sulphur dioxide

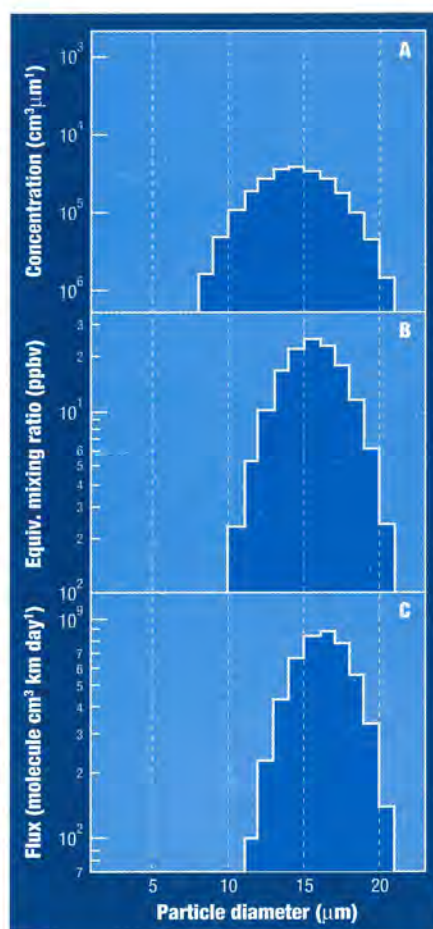


Figure 3. Results from the large PSC particle observations made by the NO_y instrument on board the NASA ER-2 aircraft. The NO_y dataset used in the analysis is an 800-s interval obtained at 19.5 km near the North Pole on January 20, 2000. A statistical simulation of the NO_y time series values was used to determine the sizes of large ($> 5 \mu m$ diameter) NAT particles sampled by the instrument. The panels show (A) the best-fit size distribution and number concentration, (B) the equivalent gas-phase mixing ratio of HNO_3 derived from the distribution, and (C) the derived sedimentation flux of HNO_3 . The concentration integral of the large mode is $2.3 \times 10^{-4} cm^{-3}$. The lower limit of the vertical axis in (A) corresponds to the detection of a single particle over the 800-s observation period. The total gas-phase HNO_3 in the large mode is 1.5 ppbv. The HNO_3 flux of the combined distribution is 5×10^9 molecules $cm^{-3} km day^{-1}$ or 2.2 ppbv $km day^{-1}$ at 60 hPa. Significant denitrification would result if this flux were maintained in the lower stratosphere for a period of a few days or more. [Adapted from Fahey et al. *Science* 2001, 291, 1026-1031].

and hydrogen peroxide, which play major role in the formation of sulphate from SO_2 in the troposphere.

These talks were followed by description of kinetics studies on OCIO, balloon borne measurements of PSCs, and modelling the effect of short-lived halocarbons on stratospheric ozone.

Solar Cycle Forcing of the Stratosphere and Troposphere workshop

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The workshop was convened to investigate the importance of solar UV variability and energetic particles in modifying stratospheric and/or tropospheric circulation on the time scale of the 11-year solar cycle. A series of invited reviews was interspersed with shorter contributed talks. Ample time for discussion was provided.

Effects of spectral irradiance changes

In a review talk **K. Labitzke** summarised the statistically derived structure of the solar cycle response in the lower stratosphere (LS) and troposphere, based on data encompassing four solar cycles. Differences of geopotential heights and temperatures between solar minimum and maximum were estimated in the LS using both reanalysed NCEP data and analyses performed at the FUB, Berlin. It was pointed out that the global solar signal in January and February is modulated by the equatorial wind QBO. **J. Staehelin** emphasised the importance of including the QBO and NAO as dependent variables in regression analyses which attempt to estimate solar cycle responses in the LS. Using the Arosa total ozone series (1926 to present), it was shown that an apparent solar cycle correlation disappears when both the QBO and the NAO are included in the regression model. Using the Payerne ozone profile series, it was also shown that the influence of the solar cycle at this location is largest in the summer season at 26 to 30 km. **L. Hood** reviewed satellite data analyses indicating a significant response of ozone and temperature to solar cycle forcing in both the upper stratosphere (US) and the LS. From SBUV data, the ozone variation in the US from solar minimum to maximum was estimated by multiple regression analysis and found to be more than 3% while that in the middle stratosphere was nearly zero. This apparent direct photochemically induced ozone response in the US is accompanied by a temperature response, with a maximum of about 0.9 K near 3 hPa (from SSU data). In the tropical LS (40 to 120 hPa), the MSU ch. 4 data indicate a solar cycle component of temperature variability of 0.70 ± 0.20 K. Based on a combined satellite total ozone data record covering 2 solar cycles, the solar cycle component

of tropical total ozone variability has an amplitude of several per cent. Most of the column ozone variation occurs in the LS. **M. Salby** reviewed statistical evidence that the equatorial wind QBO may itself vary slightly with the solar cycle. For example, based on a 40-year record, it appears that the QBO westerly phase at levels below 30 hPa tends to be longer in duration under solar minimum conditions than under solar maximum. These changes in westerly wind duration introduce a drift into the QBO phase in winter months, when the polar vortex is sensitive to equatorial wind direction. It was therefore suggested that this apparent solar modulation of the QBO may explain the large observed solar cycle variation of polar LS temperatures when data are separated according to QBO phase.

In one of a series of talks on GCM studies, **J. Haigh** discussed recent simulations of the effect of realistic solar spectral irradiance changes on the stratosphere and troposphere. Using two different GCM's, a reproducible mode of response was found in zonal mean temperature and wind fields, especially at low latitudes and in the summer hemisphere. Results were shown to be sensitive to whether solar-induced ozone changes in the stratosphere were included in the model. **K. Weber** emphasised that present GCM simulations tend to underestimate the amplitudes of observed changes or even yield contrary results. In part, these differences are due to differences in the formulation of parameterisations, such as those which determine solar-induced changes in ozone concentration. Using the University of Illinois GCM with interactive photochemistry, **E. Rozanov** obtained an ozone mixing ratio increase of up to 3.5% and a 0.5-1 K warming from solar minimum to maximum in the tropical US. A weak acceleration of the stratospheric polar night jets was also detectable. Propagation of the solar signal downward and northward during the boreal winter resulted in an intensification of the polar vortex during the late winter and early spring. The pattern of model estimated surface air temperature changes in December was similar to that observed after volcanic aerosol injections. **K. Kodera** suggested that an understanding of the diversity of dynamical responses to solar cycle forcing obtained in different models requires a consideration of the balance between solar radiative forcing and dynamical forcing due to planetary waves. In particular, differences in model parameterisations may have the largest impact

in early winter during the transition period from a radiatively controlled state to a dynamically controlled state. It is during this period when solar radiative forcing can have the greatest impact, thereby perturbing the winter stratosphere into one of its preferred internal modes, e.g., the positive mode of the Arctic Oscillation.

Effects of energetic particle precipitation

D. Siskind first discussed long-term variations in upper atmospheric nitric oxide and coupling to the middle atmosphere. It has only recently become apparent that this variability maps down into the stratosphere at polar latitudes during winter. Associated with these NO_x enhancements near solar maximum is a reduction of springtime polar ozone by up to 40% between 10 and 20 hPa. Transport of odd nitrogen to lower latitudes in the stratosphere is more controversial. However, **L. Callis** presented model simulations suggesting that non-auroral energetic electrons can contribute significantly to the odd nitrogen budget of the mid to high latitude stratosphere, thereby also affecting the solar cycle variation of ozone. He argued that this neglected contribution from energetic electron precipitation can potentially account for existing differences between observational estimates of the solar cycle ozone variation and model estimates based on solar UV variations alone. **A. Krivolutsky** conducted statistical analyses of total ozone and Galactic Cosmic Ray (GCR) fluxes at several high-latitude Russian stations. In contrast to theoretical expectations based on odd nitrogen chemistry, it was found that total ozone tends to increase during years of increased GCR fluxes. **B. Tinsley** presented a brief overview of his model for explaining possible correlations of global cloud cover and winter storm tracks with solar activity variations. The model hypothesises that solar cycle changes in GCR flux can modify the global electric circuit with indirect effects on the formation of cloud condensation nuclei (CCN). In an invited review, **J. E. Kristjansson** questioned the robustness of previously reported correlations between decadal changes in GCR flux and global cloud cover. Using alternate satellite cloud cover data, he found that the earlier correlation does not continue after 1988. He then discussed the theoretical issue of whether cosmic ray ionisation can create CCN in nature. **M. Todd** repor-

ted on a statistical investigation of the relationship between cloud cover and short-term (Forbush) decreases of GCR's. Analysis of data averaged in geomagnetic latitude bands yielded significant correlations mainly at high latitudes. The cloud/GCR correlations were found primarily using high-level cloud data (10-15 km) and were especially pronounced over Antarctica. Analysis of Forbush decrease events associated with solar proton burst events showed no statistically significant cloud anomalies.

Conclusions

Current research focuses primarily on the hypothesis that direct effects of solar UV variability in the US, especially under near-solstice conditions in early winter, can affect the selection of preferred internal modes in the northern winter stratosphere. Like volcanic forcing, solar forcing tends to favour the positive mode of the northern annular mode (or Arctic Oscillation) in the late winter LS, a mode that has been found observationally to be connected with tropospheric circulation anomalies. However, existing GCMs do not yet accurately simulate the magnitude or, in some cases, even the sense of the LS responses derived from observations. Of special interest in the area of energetic particle precipitation is the evidence summarised above for short-term cloud anomalies at high geomagnetic latitudes and altitudes associated with GCR Forbush decreases. Such evidence, if verified by later studies, would add credence to the concept that GCR fluxes can affect the production of cloud condensation nuclei. However, statistical evidence for GCR effects on cloud cover at lower latitudes and altitudes on any time scale must be categorised at present as inconclusive.

Review of AGU Fall Meeting Session: New Insights into Stratospheric Chemistry, Dynamics, and Transport

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Introduction

This session, convened by Emily Shuckburgh and Warwick Norton, presented recent advances in middle atmosphere chemistry, dynamics and transport. The session included 21 talks and 51 posters. The talks were broadly divided into four areas: (i) the effects of climate change on the stratosphere, (ii) new results and diagnostics of tracer transport, (iii) analysis of recent field campaigns and satellite measurements, with particular focus on the results of SOLVE-THESEO 2000, and (iv) understanding the distribution and transport of water vapour.

Climate change

Elisa Manzini (invited) presented results of simulations using the MA-ECHAM4-CHEM model to evaluate temperature and circulation changes related to changes in atmospheric composition from 1960 to 2000. With respect to the 1960 simulation, the 2000 simulation showed significant average cooling (~ 6 K) in March [30–10 hPa], strengthening of the zonal winds (~ 8 m/s), and local decrease ($\sim 10\%$) in downwelling, mainly due to a change in the contribution from planetary waves. In contrast, in the mesosphere, downwelling is enhanced ($\sim 20\%$); the contribution from gravity waves is twice that from planetary waves.

Christoph Brühl showed that the MA-ECHAM4-CHEM model, which includes interactive photochemistry, could reproduce the main features of global temperature and ozone distributions in the sixties and the nineties.

David Rind used the GISS Global Climate/Middle Atmosphere Model to examine the effect on the stratospheric circulation and tracer transport of doubled CO_2 and two alternative sets of SSTs, one with greater low latitude and overall warming. While both results in an intensified subtropical circulation in the lower stratosphere (LS), they have opposite effects on the high latitude residual circulation and the Arctic Oscillation phase.

Piers Forster (invited) used the latest SPARC water vapour trends (assuming the NH mid-latitude trend to be representative of the global trend) to model

stratospheric temperature changes and to calculate radiative forcing. He finds significant sensitivity in the model response to the imposed background water vapour concentration, but, with the best-guess background and trend applied in model, he obtains a 0.8 K global cooling of LS for the 1980–2000 trend and a 1.8 K cooling for 1960–2000 trend, equating to radiative forcings of 0.3 Wm^{-2} over last 20 years or 0.6 over last 40 years. These forcings are large (75% of the CO_2 forcing or 40% of the well-mixed greenhouse gas forcing) and hence potentially important.

Betsy Weatherhead discussed optimal methods of detecting the gradual recovery in the total column ozone record, expected to be only a few Dobson Units per decade over the next 50 years. She explained that variations in the seasonal and altitudinal distributions of ozone suggest that evidence of recovery may be easier to detect during the local springtime, or at a single vertical layer in the atmosphere.

John Farrara used the UCLA GCM to examine the response to reduced stratospheric ozone, which may arise from reduced solar activity. He suggested an amplification mechanism, which through coupling by stratospheric annular modes could contribute to the little ice age period.

Tracer transport

Bob Hudson (invited) showed that the total ozone distribution can be separated into four “meteorological regimes” divided by the subtropical upper troposphere (UT) front; the polar front and the polar vortex, with the distribution often being rather narrow in the first three, mid-latitude, regimes. He showed that the mean total ozone value within each of these mid-latitude regimes is relatively constant from year to year, suggesting that it is the relative areas of the three regimes that determine the mean mid-latitude (20 to 60°N) total ozone. He proposed that most of the northern mid-latitude ozone trend between 1978 and 1992 is in fact a consequence of a northward movement of the mean latitude of both the subtropical and polar fronts.

Anne Douglass examined diagnostics of tropical transport in CTM simulations driven by winds from the Finite Volume CCM and the DAO analyses. Signatures of interannual differences related to the tropical winds and extratropical waves were presented to understand differences between the two model transports and the transport inferred from the observations.

Fred Moore used *in situ* measurements of long-lived trace gases to calculate a distribution of maximum path heights over which air has travelled in the stratosphere. The measured maximum path height distribution is a transport diagnostic, which is independent of measured mean age or age distributions and thus provides an additional check on model derived stratospheric transport.

Bernard Legras demonstrated how ideas and concepts from the theory of dynamical systems could be used to investigate the transport characteristics of the stratospheric polar vortex. He used “finite-size Lyapunov exponents” to identify so-called stable and unstable manifolds in the vicinity of the vortex, and showed how these structures are associated with transport of particles.

Field campaigns and satellite measurements

Megan Northway showed results from SOLVE-THESEO, which indicate widespread denitrification near 40% between 20–30 km in the polar vortex. To explain this, particle populations must have molecular HNO_3 flux values between 10^9 and 10^{10} molecules $\text{cm}^{-3} \text{ km day}^{-1}$, these flux values must be achieved near 20 km for a total of several days, particle sizes need to be 5–20 μm diameter, and number concentrations less than 0.1 cm^{-3} .

Paul Wennberg (invited) and co-authors illustrated how the well-known formation of NAT particles in localised lee-wave mountain ice clouds can produce a low number density of large particles in the LS. They showed, using a simplified model, that this mechanism for nucleation of NAT particles is quite consistent with observations made during SOLVE-THESEO. Other more speculative nucleation mechanisms (e.g. selective homogeneous or heterogeneous

nucleation) may not be required to explain denitrification of the polar vortex. [Manuscript now in press at GRL, Dhaniyala, McKinney, and Wennberg, 2002].

Steve Eckerman explained that global forecast models used during SOLVE-THESEO indicated that the very coldest intravortex regions were small "meso-scale cold pools" occurring recurrently over the East Coast of Greenland, embedded within a broader synoptic cold pool region. He showed, based on archived forecasts and analyses from various global models, that these features are not artefacts of sigma coordinate models induced by the Greenland coastal topography, but are produced by a long-wavelength mountain waves, launched by eastward flow over the Greenland ice shelf.

Paul Konopka presented the results of high-resolution ClAMS model simulations of the chemical composition of the Arctic stratosphere for late spring 2000, and discussed the impact of mixing on ozone depletion. He showed that the deactivation of ClO_x within the air masses originating from the polar vortex occurred, even after the vortex break-up, mainly through the photochemical NO_x production from HNO_3 without a significant contribution from mixing with NO_x -rich mid-latitude air. He also showed the mid-latitude ozone deficit in late spring was dominated by dilution by polar air and, until mid of April, by ClO_x -induced ozone destruction within the vortex remnants.

Hideaki Nakajima presented tracer data from the Improved Limb Atmospheric Spectrometer (ILAS) on board the ADEOS satellite. When plotted in equivalent latitude space, the data nicely showed the vortex edge and subtropical barriers.

Peter Preusse analysed the high spatial resolution stratospheric temperature data sampled by CRISTA for gravity waves. Deduced gravity wave amplitudes were correlated to a tropopause-height/convection proxy inferred from CRISTA water vapour and cloud top heights. This was thought to be due to two reasons: deep convection directly excites gravity waves as illustrated by super typhoon Winnie, and the vanishing Coriolis force at the equator allows the existence of low frequency gravity waves with very long horizontal wavelengths enhancing gravity wave energy in the tropics.

Water vapour

Steve Sherwood (invited) showed there exists a close coherence between relative humidity variations just below the tropical tropopause, and moisture variations later at higher levels, from which temperature effects have been removed. He demonstrated that there is a highly significant correlation between the relative humidity of air crossing the tropopause and the effective diameter of ice crystals in cumulonimbus cloud tops, consistent with a physical explanation based on crystal fall speeds and evaporation rates. There is a likely link of crystal sizes to droplet nucleation by biomass burning particles at earlier cloud stages, and this is a likely contributor to the upward trends in stratospheric water, however, absolute calibration of trends in crystal diameter appears impossible at this time.

Koji Yamazaki used an AGCM simulation forced by observed SSTs to examine the downward time-mean vertical motion near the tropopause over Indonesia. A heat budget calculation showed that cold advection from the east and reduced longwave heating balances the warming by downward motion.

Sensitivity calculations of the cloud height and temperature profiles showed that the high cloud suppresses the long-wave radiative warming around the tropopause region.

Daniel Kirk-Davidoff presented preliminary results from the NASA-funded CWVCS airborne field mission, based in Costa Rica, to study the mechanisms controlling the distribution and transport of water vapour in the tropical upper tropopause and LS. Measurements were made, including ozone, water vapour, total water and methane, in and far from regions heavily influenced by deep convection. A wide variety of very low-temperature cirrus clouds were intercepted, and tropopause temperatures as low as 185 K were encountered.

Zhiming Kuang showed observations of the isotopic ratio of water vapour in the tropical UT and LS from ATMOS and the MkIV balloon flights, and noted that the isotopic ratios were not inconsistent with dehydration due to condensation down to water vapour mixing ratios of roughly 10 ppmv. Further drying of air down to 3-5 ppmv, which is typical of air entering the tropical stratosphere, was postulated to be due to some form of mixing, since the isotopic ratios were inconsistent with condensation as the exclusive drying mechanism below 10 ppmv.

Ghassan Taha explained that recent improvements to the SAGE II retrieval algorithm have resulted in enhancements in the vertical structure of the observed profiles. He discussed the latitudinal distribution and the climatology/interannual variation of water vapour obtained, and compared these to measurements from HALOE and MLS.



School on the Equatorial Middle Atmosphere and Ionosphere

24 September-5 October 2001, Trieste, Italy

Kevin Hamilton, University of Hawaii, Honolulu, USA (kph@soest.hawaii.edu)

A School on the "Physics of the Equatorial Atmosphere" was held September 24-October 5 at the Abdus Salam International Centre for Theoretical Physics (ICTP) in Trieste, Italy. The ICTP is sponsored by the United Nations Educational, Scientific, and Cultural Organisation (UNESCO) and is devoted to facilitating graduate

level education and research opportunities for scientists from developing countries. The Equatorial Physics School was co-sponsored by SPARC and it reviewed the current state of research on a range of aspects of the low-latitude atmosphere including meteorological phenomena as well as phenomena related to stratospheric

ozone chemistry and ionospheric physics. The School attracted 42 students from 27 countries, including 12 students from Africa, 8 from South Asia and Indonesia, and 8 from Latin America. Lectures were given by 11 distinguished scientists from the USA, Japan, the UK, Germany, Australia, India and Brazil.

Future SPARC and SPARC-related Meetings

- 18-22 March 2002: International Symposium on Equatorial Processes Including Coupling (EPIC)**, Kyoto University, Uji, Kyoto, Japan. Conveners: S. Fukao, J. Forbes, K. Hamilton, R. Vincent (epicsympo@kurasc.kyoto-u.ac.jp).
- 22-26 April 2002: EGS XXVII General Assembly**, Nice, France.
(<http://www.copernicus.org/EGS/egsga/nice02/programme/overview.htm>).
- OA20:** Tropospheric chemistry and aerosols. Co-conveners: M. Ammann, F. Dentener, Ch. George (christian.george@univ-lyon1.fr), T. Hoffmann, T. Koop.
- OA21:** Dynamics and chemistry of the UT/LS. Convener: V. Wirth (vwirth@mail.uni-mainz.de).
- OA23.02:** Climate variability: The northern oscillation: NAO and AO. Convener: U. Ulbrich (ulbrich@meteo.uni-koeln.de).
- ST3:** Open session on the middle atmosphere. Convener: M. Juckes (juckes@atm.ox.ac.uk).
- ST4:** Atmospheric ozone (conveners: P. Fabian (fabian@met.forst.uni-muenchen.de), G.O. Braathen, P. Taalas, A. Volz-Thomas).
- ST5:** New developments in stratospheric and mesospheric chemistry. Convener: J.M. Plane (j.plane@uea.ac.uk).
- ST7:** Impact of aircraft emissions on climate and atmospheric chemistry. Convener: R. Sausen (sausen@dlr.de).
- ST8:** Chemistry-climate interaction. Convener: M. Dameris (martin.dameris@dlr.de).
- 13-16 May 2002: 276. WE-Heraeus-Seminar on Trends in the Upper Atmosphere**, (upon invitation only), IAP Kühlungsborn Germany (http://www.iap-kborn.de/heraeus/index_d.htm). Convener: F. J. Lübken (luebken@iap-kborn.de)
- 19 May-01 June 2002: NATO Advanced Study Institute (NASI) on Data Assimilation for the Earth Sciences**, Acquafredda di Maratea, Italy. Co-chairs: R. Swinbank (richard.swinbank@metoffice.com), V. Shutyaev (shutyaev@inm.ras.ru).
- 27-30 May 2002: GAW Workshop for WMO Region VI (Europe) on Status and Trends of Global Atmosphere Watch with particular emphasis on the situation and needs of WMO RA VI (Europe)**, Riga, Latvia (joerg.klausen@empa.ch).
- 08-12 July 2002: Fifth UT/LS Ozone Science Meeting**, Bristol, UK. Rebecca Penkett (manager@utls.nerc.ac.uk) (<http://utls.nerc.ac.uk/Meetings/Future/UTLS/sci5.html>).
- 02-06 September 2002: Sixth European Symposium on Stratospheric Ozone**, Chalmers University of Technology, Göteborg, Sweden (<http://www.ozone-sec.ch.cam.ac.uk/workshop/Goteborg/Goteborg.html>).
- 18-25 September 2002: Atmospheric Chemistry in the Earth System: from Regional Pollution to Global Change**, Crete, Greece (<http://atlas.chemistry.uoc.gr/IGAC2002>).
- 10th Scientific Conference of the IAMAS Commission for Atmospheric Chemistry and Global Pollution (CACGP) and 7th Scientific Conference of the International Global Atmospheric Chemistry Project (IGAC).**
- 10-19 October 2002: 34th COSPAR Scientific Assembly and 2nd World Space Congress**, Houston, TX, USA. (<http://www.copernicus.org/COSPAR/COSPAR.html>). Chair: S. S. Holt.
- A1.1/C2.7:** Climate Change Processes in the Stratosphere and at the Tropopause. Convener: J. Gille (gille@ncar.ucar.edu).
- C2.3:** Long-term Trends in Stratosphere-Mesosphere-Thermosphere Coupling. Convener: D.K. Chakrabarty (chakra@prl.ernet.in).
- D2.3/E3.3:** Solar Variability and Climate Changes. Convener: J.M. Pap (papj@marta.gsfc.nasa.gov)

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