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STRATOSPHERIC PROCESSES AND THEIR ROLE IN CLIMATE
A Project of the World Climate Research Programme

WCRP

The 27th Session of WCRP's Joint Steering Committee

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The 27th session of the World Climate Research Programme's (WCRP) Joint Steering Committee (JSC) was held in Pune, India, 6-11 March 2006. Attending on behalf of SPARC were the co-chairs, **Alan O'Neill** and **A. R. Ravishankara (Ravi)**, and the director of the SPARC Office, **Norm McFarlane**. On this occasion, representatives of two of WCRP's funding agencies were in attendance: delegates from the International Council of Science and the World Meteorological Organisation.

JSC meetings are the occasions when progress with WCRP projects are discussed, when connections between the different projects are highlighted or stimulated, and when the strategic direction of WCRP is examined, as are links with other international research programmes. Of particular note at the moment are the links with the International Geosphere Biosphere Programme (IGBP), concerning which SPARC is taking a leading role on behalf of WCRP in the area of atmospheric chemistry and climate.

The JSC meeting was preceded by a joint meeting of the steering committees and project leaders of WCRP and IGBP. The aim was to stimulate joint activities, such as the project on atmospheric chemistry and climate led by SPARC for WCRP and by IGAC for IGBP. Ravishankara and **Phil Rasch** (IGAC co-chair) gave a joint presentation on progress with this collaboration, a summary of which is given in this newsletter.

The JSC meeting itself got off to a lively start when the recently appointed Director of WCRP, **Ann Henderson-Sellers**, presented her vision for the future. She wished to increase the visibility of WCRP internationally, and prompted co-chairs of WCRP projects such as SPARC to consider how this might be achieved. She advocated that projects should focus on clear deliverables (as SPARC has always done), a point that stimulated some debate. The SPARC co-chairs expressed the following personal views on this matter. The prime importance of WCRP is as an international arbiter of good taste in the climate sciences. Acceptance and encouragement of new initiatives by WCRP can be an important stamp of approval for both national and international funding agencies. This role far outweighs in importance the limited funds that WCRP can allocate to its projects. Concerning deliverables, the SPARC co-chairs felt that clarity is needed about ownership. WCRP funding provides the "glue" to facilitate and enable collaborations. It is deliverables connected to this enabling function that can be attributed to WCRP, as distinct from the achievements of individual research groups, to which the agencies that funded the research would wish to lay claim. By

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the end of the meeting, discussion on these points still had some way to run, but it was agreed that the new Director had stimulated and challenged the WCRP projects to rethink constructively their modus operandi.

The scientific discussions revolved around cross-cutting themes in WCRP, reports by the modelling and observations panels, as well as reports by individual projects and working groups. The theme of anthropogenic climate change (ACC) remains a focal point of WCRP, and each of the projects was invited to comment on its contributions to this theme. On behalf of SPARC, Alan O'Neill and Norm McFarlane highlighted a number of areas where the stratosphere is relevant to anthropogenic climate change. These included the interaction of ozone (more generally, radiatively active chemical species and stratospheric aerosols) with climate, the dynamical effects of the stratosphere on the troposphere, the impact of solar variability on the upper atmosphere and then possibly on surface climate, and stratospheric changes as part of the "fingerprint" of climate change. As ACC was the cross-cutting issue that was most discussed, the main points and conclusions of this discussion are summarised in a separate short article in this issue of the newsletter.

The SPARC Presentation

Alan O'Neill gave the SPARC report. SPARC's approach on these occasions is to try to engage the JSC in discussions of issues that require their attention and advice, referring the JSC to written submissions for progress reports. Since atmospheric chemistry and climate had been discussed at length in the WCRP/IGBP session, the SPARC report focused on a single topic that is central to SPARC's mission: understanding dynamical variability in the stratosphere. The word "dynamical" here is meant to imply variability for which two-way coupling with atmospheric chemistry is hypothesised (at least provisionally) not to be central to the features of concern, so that, for example, long-term trends in chemical species would be prescribed in numerical simulations. Some of the related dynamical and modelling issues are: (1) understanding trends (and their statistical significance) in the context of natural variability; (2) determining what is needed to simulate a realistic quasi-biennial oscillation in atmospheric general circulation models; (3) determining which properties the models must have to replicate the observed age of air spectrum in the stratosphere. These and other dynamical issues have significant impacts on chemical models through temperature-dependent chemical reactions, and through the transport of chemical species. The modelling strategy to address these issues is envisaged to include experiments with simplified (hypothesis-testing) models as well as AMIP-style simulations, and as such complements well the modelling strategy adopted in SPARC's CCMVal project on evaluating, through a process-oriented approach, coupled chemistry-climate models.

Alan O'Neill reported that plans are now being formulated to move this activity forward as a natural follow-on to SPARC's GRIPS project (GCM Reality Intercomparison Project for SPARC). The GRIPS project was a pioneering activity to get a general sense of how general circulation models with a representation of the stratosphere simulated salient dynamical features, such as sudden warmings. Contributing experiments were not constrained sufficiently to facilitate the more stringent and revealing model comparisons envisaged now. The new class of experiments will be designed more precisely to allow "apples to be compared with apples". They will also recognise (*e.g.* through the use of ensembles of multi-annual simulations) the role of statistical uncertainty in limiting confidence in model intercomparisons.

A long abiding, overarching scientific question posed to SPARC by the climate modelling and prediction community is this: how well does the stratosphere need to be represented in climate models to predict climate change (usually tacitly assumed to mean climate change at the surface)? Many global atmospheric models now have a well-resolved stratosphere and an upper boundary well above the stratopause, opening the way to attack this question directly, and for SPARC to coordinate multi-institutional efforts to do so. The procedure in outline is clear enough: examine the impact on climate features or processes of degrading stratospheric resolution in comparison with control runs at full resolution, ensuring (by ensemble or equivalent means) that results are statistically significant. The JSC strongly endorsed this activity, commenting that it was timely and long overdue. With this encouragement, the new initiative on dynamical variability in the stratosphere will be a major topic for discussion and detailed planning at the next meeting of SPARC's Scientific Steering Group, to be hosted by A. R. Ravishankara in Boulder, October 2006.

Cross-cutting activities

The Atmospheric Chemistry and Climate (AC&C) activity is one of a range of activities that involve collaboration between SPARC, IGAC, and other core projects within the WCRP. A new initiative is the joint SPARC-GEWEX-IGAC activity on the role of deep convection in the Tropical Tropopause Layer (TTL). This activity will be initiated with the upcoming workshop: Modelling of Deep Convection and of Chemistry and their Roles in the Tropical Tropopause Layer, which will be held in Victoria, BC, Canada in the period of June 12-16, 2006. These activities are also relevant to the Earth System Analysis and Modelling activity of the Earth System Science Partnership (ESSP). This is a partnership of four global change programmes, WCRP, IGBP, IHDP and DIVERSITAS. (Further information can be obtained from http://www.essp.org/). The session on Chemistry and Climate of the Upper Troposphere-Lower Stratosphere (UTLS) Region at the forthcoming ESSP Open Science Conference (http://www.essp.org/essp/ESSP2006/) will provide a forum for presentation of key components of the research underpinning the AC&C activity.

In addition to anthropogenic climate change, monsoons and extreme events were selected for discussion as cross-cutting topics at the JSC meeting. The first Pan-WCRP Monsoon workshop was held at the University of California, Irvine (USA) in June, 2005. (See http://www.wmo.ch/web/wcrp/wcrp_informal_reports.htm for a full report). The objectives of this workshop were to assess current understanding of the basic physical processes governing various monsoon climate systems, to facilitate better predictions using a hierarchy of models, and to promote a pan-WCRP initiative on monsoon prediction studies as a component of the COPES programme. Findings and recommendations to the JSC were put forward in a presentation by **T. Yasunari** on behalf of the organizers. Prediction is a challenging problem for current models because monsoon sys-

tems consist of processes and variability that operate on a wide range of spatial and temporal scales. Among current prediction model shortcomings are inaccurate modelling of the diurnal cycle of precipitation, and an inability to realistically model the structure, amplitude, and frequencies of intraseasonal oscillations and their coupling to convective activity. A better understanding of moisture transport processes and of the spatial resolution is needed to adequately simulate the multi-scale interactions that dominate monsoon systems.

The workshop was jointly sponsored and organised by GEWEX and CLIVAR, both of which have monsoon panels and ongoing activities which explicitly focus on monsoon systems. The JSC recommended that the CLIVAR and GEWEX monsoon panels should continue to work closely together and (with participation of SPARC and CliC, and JSC representatives) establish focal points to facilitate development of a coordinated programme on monsoon research that will be presented for discussion at the next JSC meeting. WCRP Modelling Panel (WMP) should be prepared to coordinate the relevant modelling activities with participation of SPARC and Coordinated Enhanced Observing Period (CEOP). Participation of WGNE and THORPEX in these activities is also very important, particularly in regard to the focus on improved modelling of the diurnal cycle.

Information about extreme weather events and how they may change in association with climate change is increasingly in demand. If climate variability can be characterised by probability distributions of weather events, then extreme events are those that are characterised by the tails of the distribution functions and may therefore be intrinsically difficult to predict. This topic is nascent and increasingly important. A recommendation arising from the discussion at the JSC meeting is that the WCRP should set up a framework for studying extreme events to address data, modelling, simulation and predictability needs. An initial step is a session on extremes at the seasonal prediction workshop in 2007. The JSC will continue to address the cross-cutting between extremes and ACC in its future sessions. Although the discussion largely focused on surface manifestations of extreme events, extremes in the stratospheric circulation (*e.g.* unusually cold and intense Arctic winter polar vortices, or rare Antarctic mid-winter warmings) affect the seasonal distribution and evolution of ozone and may also affect surface meteorological conditions through stratosphere-troposphere dynamical coupling. Therefore, this is also a topic of importance for SPARC.

Activities in WCRP panels and working groups and links to other programmes

Modelling is now central to many of the ESSP activities. The goal of the ESSP Modelling Strategy is to encourage modelling activities that will address the range of issues that are the concern of partnership members. These include a broader range of components (carbon cycle, dynamic vegetation, biogeochemical cycles, coupling between human and natural systems), and time scales than have typically been dealt with hitherto in weather and climate prediction models. This involves using a variety of modelling strategies and configurations. GCMs and NWP models are central to activities that fall within the purview of the WCRP, consistent with the emphasis on assimilation and prediction in the COPES framework, while EMICS (Earth Systems Models of Intermediate Complexity) and mechanistic models of coupled physical, chemical, biological, and human systems have been more prominent in the other programmes within the ESSP.

Within the WCRP, modelling is prominent in the programmes of core projects (SPARC, CLIVAR, GEWEX), the activities of the Task Force on Seasonal Prediction (TF-SP) and all of the cross-cutting topics of current concern. Its central role is the main reason for the existence of the modelling working groups and panels (WMP, WGCM, WGNE). Many of the activities being dealt with in these panels involve coordination with other programmes, such as THORPEX.

The current activities of the TF-SP are directed toward determining the extent to which global prediction and validation on seasonal time scales are possible with currently available models and data, identifying limitations of current models and data in determining seasonal predictability, and developing a plan for Climate System retrospective seasonal forecasting experiments. For example, this involves using and comparing high resolution NWP models and modest resolution IPCC class models for seasonal prediction. The importance of stratospheric processes in seasonal prediction is an abiding issue for SPARC. This issue is being addressed within the SPARC theme of detection and prediction and the new dynamical modelling activity will enhance the ability of SPARC to contribute to the goals of the TF-SP.

The role of the Working Group on Numerical Experimentation (WGNE) is to support the modelling community by addressing fundamental issues in NWP and climate modelling such as dynamical cores, resolution and representation of physical processes. For example, routine monitoring of the skills of daily forecasts from the major operational centres has provided a useful indication of improvements and remaining deficiencies in forecast systems. For climate modelling, WGNE activities are coordinated with activities of the Working Group on Coupled Modelling (WGCM). The intercomparisons AMIP I, AMIP II and the follow on CMIP intercomparision (*e.g.* of the IPCC AR4 climate models) have encouraged and revealed improvements in a number of areas, such as the representation of stratus and stratocumulus cloud systems. However, a number of significant deficiencies remain as issues for both NWP and climate models. Prominent among these are abiding deficiencies in representing the spatial structure and temporal variability of convective precipitation. As noted above in connection with monsoon prediction, these shortcomings are manifest in deficiencies in simulation of the diurnal cycle of convective rainfall. They are also central to deficiencies in simulation of intra-seasonal oscillations in the troposphere such as the Madden-Julian Oscillation. For SPARC understanding and modelling, the role of deep convection in the forcing of the Quasi-Bienneial Oscillation and the Semi-Annual Oscillation are also abiding issues that are related to these same deficiencies in modelling of the effects on convection in GCMs.

Because of these issues and the importance of convection in both NWP and Climate prediction, the JSC has endorsed a proposal by the WGNE for a coordinated WCRP effort on convection. An important aspect of this proposal involves carrying out a small number (because of the associ-

ated computational expense) of very high resolution (~ 1 km in the horizontal) forecasts/simulations over large domains using comprehensive models to study the interaction of moist physics and the large-scale environment, and to provide input to, and constraints on, improving parameterizations. These high resolution simulations would be accompanied by a number of lower resolution simulations with global models, providing a context for studying resolution-related issues such as the convergence of parameterizations. The proposed SPARC dynamical modelling activity would be complementary to this proposal.

In addition to addressing and quantifying uncertainties in climate prediction, and reducing those associated with systematic modelling errors, the WGCM also addresses a range of other issues in climate prediction. These include following up on issues raised by the IPCC and anticipating IPCC needs, addressing issues of long term variability and the role of historical radiative forcing variations in detection and attribution of climate change. Many of these activities are complementary to corresponding parts of the SPARC programme.

The mandate of the WMP is to promote and coordinate modelling activities across the WCRP with the goal of meeting the objectives of COPES. Consequently, its membership includes representatives from all of the WCRP projects and working groups as well as from other partner programmes in ESSP and the THORPEX programme. Because it was constituted relatively recently, this panel met for the first time in the past year. At this meeting a range of scientific questions and issues were put forward for ongoing consideration of the panel. These include seamless prediction of weather and climate, initialization for daily, seasonal, and decadal prediction, the importance of resolution (*e.g.* cyclone resolving *vs.* cloud resolving) and associated computational requirements and constraints (short term runs with high resolution models, long term runs with lower resolution models, ensembles, periods of integration), data issues and trends in computing. Many of these issues overlap with those of the THORPEX programme. A "white paper" is being produced jointly by WMP and THORPEX to facilitate a coordinated approach to the range of common issues. These include high resolution global weather and climate prediction, data assimilation, design and implementation of an Earth Observing System that will address future needs of weather and climate prediction, contribute to advances in prediction of weather hazards, extreme events and their inherent uncertainty, and climate variability and change.

The WCRP Observations and Assimilation Panel (WOAP) was established to complement the modelling panel (WMP) by fostering and promoting syntheses of observations. The first meeting of WOAP was held in June, 2005. The terms of reference for WOAP are to identify climate observational requirements, help to optimise observations, to act as a focal point for WCRP interactions with other observational groups and activities, to promote and coordinate analysis, reprocessing, reanalysis and assimilation activities, to promote and coordinate information and data management activities. An initial focus of WOAP will be on reanalysis with the goal of addressing the need for internationally coordinated, enhanced, and sustained reanalysis activities to meet the needs of climate monitoring, establish ocean reanalysis for the recent satellite era, and include variables related to atmospheric composition and climate forcing. These issues will be addressed initially in a reanalysis workshop to be held at ECMWF in June, 2006 and a further in a major reanalysis conference in Japan late in 2007.

As a core WCRP project SPARC is represented on WMP and WOAP, and maintains ongoing contacts with WGNE and WGCM. These interactions are complementary to ongoing collaborations between SPARC and the other core WCRP projects, particularly with CLIVAR and GEWEX. They are increasingly important for connecting the SPARC programme to other activities such as those of the IPCC as well as to the objectives of COPES.

Atmospheric Chemistry and Climate

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All human-produced radiatively active gases and aerosols are chemically active, with the lone exception of carbon dioxide, in the atmosphere. In all these cases, the removal of the forcing agents from the atmosphere is controlled by or influenced by atmospheric chemical processes. Chemical processes both in the stratosphere and in the troposphere dictate the production of some ozone, a key radiatively active gas. Thus, the abundances of climate forcing agents are critically dependent on the chemistry that occurs in the atmosphere. Further, it is precisely these chemically active agents that are short enough lived to be influenced by human action on a short time scale. Also, the future forcing, as well as the current forcing, of the short-lived agents have to be calculated and are not based on their observed abundances in the atmosphere, which varies a great deal spatially and temporally. Because of these reasons, it is clear that good representation of the chemical processes in climate and earth system models is essential.

The genesis of Atmospheric Chemistry and Climate is the recognition that *both* IGBP and WCRP recognise that this topic is important and it is time to bring together relevant research taking place under the umbrellas of the two organizations.

SPARC, a project of WCRP, has focused heavily on the modelling and understanding of the stratosphere in general and stratospheric ozone in particular. IGAC, a project of IGBP has focused on understanding the processes that occur in the troposphere. Therefore, the gap of modelling tropospheric chemistry in climate and earth system models has not been the major purview of either IGBP or WCRP. Of course, modelling centres and communities have necessarily included tropospheric chemistry in their models. It is, therefore, clear that a "coordinating and bringing together" role of international umbrella organizations such WCRP and IGBP is needed. To fulfill this need, WCRP and IGBP asked SPARC and IGAC to come together to strategise and develop a road map for a joint WCRP-IGBP project or activity to address this issue.

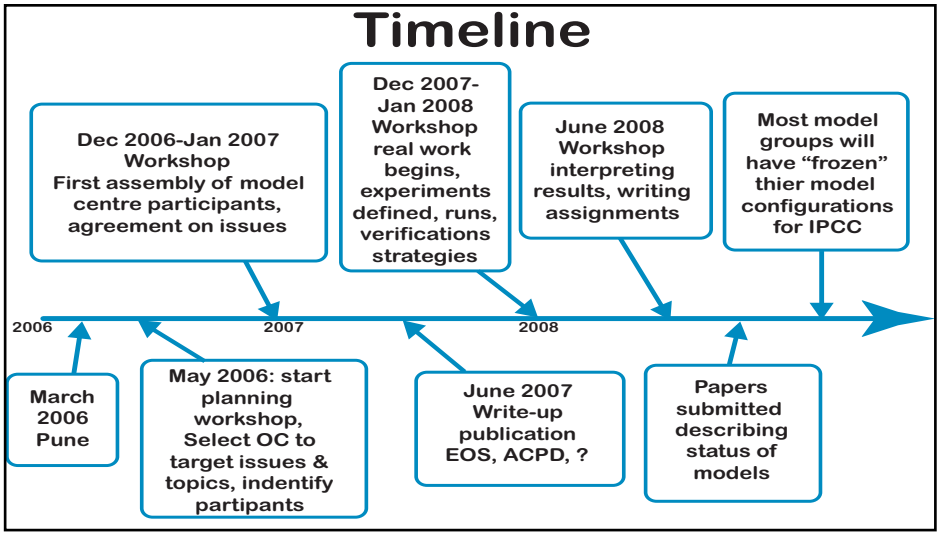
A.R. Ravishankara from SPARC and **Phil Rasch** from IGAC were asked to lead the development of this roadmap and have been working on its development. After mutual consultations and talking to other scientists, they have a skeleton of a plan to flesh out this project. In defining this roadmap, the initial ideas were presented to the SSG of SPARC and SSG of IGAC. The comments and other input from the members helped shape the focus and approach. Some excellent ideas presented were put on "hold" to get the project started. They presented this plan in the WCRP-IGBP joint session at Pune.

WCRP and IGBP have previously made connections in some areas of research, but always at a "high" level; these connections seldom came down to the level of involvement of projects of each organization. However, one of the interesting developments was the joint SPARC-IGAC activity that was initiated a few years back, in the area of laboratory studies. Then, the Giens workshop, which took the first step towards exploring the current state of atmospheric chemistry and climate, started a more in-depth interaction. These joint activities, and more importantly the fact that the "same" scientists and institutions are involved in both SPARC and IGAC, makes this joint venture natural, comfortable, and easy to initiate. Further, the complementary nature of IGAC and SPARC make it even more attractive.

The initial phase of this map involves a focus on modelling for the following reasons: (1) The observations, key areas for IGAC, SPARC, and many other units within IGBP and WCRP, provide insight into the processes and their representation in models. (2) Detailed component modelling studies provide the needed realistic representation of how a process works and a way to incorporate them in the global models. They still remain the emphases of the participating core projects. (3) The large-scale global models will be the purview of the new venture, with a heavy reliance on the core projects for the components. The large-scale models then will allow exploration of policy relevant issues using models that are observationally and physically vetted. (4) Core projects already have some modeling activities, *e.g.*, CCMVal in SPARC, that can be extended to the troposphere.

With these thoughts in mind, it was proposed that the AC&C concentrate on modelling in global models to explore the issues of (a) aerosols, (b) tropospheric ozone, and c) deposition processes. The first two are key societal issues in climate and its impact. The third is a process that has remained a "parameterization" and could use more "physical science" oriented approach.

The time line for the roadmap has been developed and is shown in the accompanying figure. It consists initially of a series of workshops to define the specific targets and quickly define a set of modelling experiments that are aimed towards providing some key answers to the next round of assessments.



Anthropogenic Climate Change

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As noted in an accompanying report in this newsletter, anthropogenic climate change (ACC) was selected as one of the cross-cutting issues for focused discussion at the WCRP JSC meeting in Pune. With both the IPCC AR4 and WMO/UNEP Ozone assessments nearing completion, it is timely to consider what has been learned from these assessment processes. The COPES framework provides the context for the WCRP to contribute more effectively and critically to future assessments by fostering and facilitating the research needed to reduce remaining uncertainties in detection, attribution, and prediction of ACC.

In the SPARC presentation on ACC we noted that the cross-cutting issues of atmospheric chemistry and climate, and anthropogenic climate change are closely linked and are prominent in the SPARC programme. The SPARC presentation focused on ozone depletion and recovery, and detection and attribution of stratospheric changes.

Stratospheric Ozone depletion and its recovery: What to expect and what are the needs?

Stratospheric ozone depletion is a major environmental issue that has been recognised and diagnosed. Based on the scientific findings and predictions, policy decisions have been made to eliminate or restrict ozone-depleting substances (ODS) *via* the Montreal Protocol and its many amendments. The "recovery" of stratospheric ozone levels to pre-chlorofluorocarbon levels in response to the Montreal Protocol is expected in the coming decades, with the abundances of ODS peaking during the present decade. Detecting the early signs of the recovery, monitoring the ozone layer through the transition period, and finally seeing it through the consequences of the protocol are issues of major societal importance.

The influence of changing climate is foremost among the factors affecting the ozone layer, its depletion and recovery. While ODS amounts in the atmosphere have peaked and are slowly decreasing, the climate of the stratosphere is also changing. Therefore, correct attribution of the ozone changes to policy actions on ODS, as opposed to changes in stratospheric climate, is a key need. Also, predicting how climate change hastens or delays recovery is an important issue. A second important issue is how the changes in the tropospheric abundances of ODS translate into changes in the ozone-depleting active chemicals in the stratosphere. Dynamical processes that control the transport and mixing time scales involved need to be quantified. In addition, dynamical issues related to vortex formation and sustenance need to be carefully taken into consideration, especially when predicting the future of the polar ozone. Such a prediction is important because of societal concerns. The importance of naturally occurring short-lived halogen containing species on the abundance of active halogens in the stratosphere has to be carefully assessed. Such an assessment is key for the possible introduction of short-lived species as substitutes for the longer lived ODSs, as well as for making decisions on current exemptions of some chemicals. The influence of the changes in stratospheric ozone on the Earth's climate needs evaluation, since these changes in the stratosphere can also influence the composition of the troposphere. For instance, intrusion of stratospheric ozone to the ground level can influence the air quality at the surface.

In addition to the scientific needs and issues noted above, the very task of assessments of the ozone layer, required by the Montreal Protocol every four years, relies on the "footwork" of organizations such as WCRP and IGBP through their projects. This work provides the basis for the assessments and brings together the pool of scientists that carry out the assessments.

Attribution of stratospheric changes, role of natural variability and modelling

One of the prominent issues that arises in attributing climate change in the stratosphere is accounting for the natural variability in the coupled system. Modelling and attribution of the changes to specific effects are important. In this regard, two broad issues have emerged:

Stratospheric change as part of attribution and detection of climate change: If stratospheric changes are anthropogenic and not due to natural variability, we should expect our models to reproduce the changes, and to the extent that they do, we gain confidence in our models and in our understanding of the relevant processes. A key question is how to characterise the statistics of natural variability. This is particularly challenging for the Arctic stratosphere where both observations and model results show a great deal of long-term variability, not just from year-to-year but on decadal time scales. This is similar to the time scale of the changes in ODS forcing, and is also the length of the satellite record. Thus, for example, although chemical ozone loss attributable to ODSs occurs in the Arctic and can be very considerable in anomalously cold winters, this loss is strongly modulated by dynamical variability, which can also affect ozone transport. The two effects may act in concert, with years with low transport being associated with lower temperatures and enhanced chemical ozone loss. Thus the Arctic stratosphere ozone layer is potentially sensitive to changes in temperature and transport in response to changes in planetary wave forcing/dissipation, sometimes referred to as planetary wave drag (PWD), associated with ACC. The comparison of Austin *et al*. (2003) indicates considerable scatter in model results in this regard, with some predicting an increase and some a decrease of PWD over the next several decades resulting from climate change. These

differences may reflect differences between the models, but they may also reflect insufficient statistical sampling. A major modelling challenge is to characterise the variability statistics sufficiently well to enable detection and attribution of ACC in the stratosphere.

Changes in the stratosphere affecting meteorological (dynamical) variability in the troposphere: There is evidence that the state of the stratosphere can have an impact on the troposphere on a variety of time scales (medium-range to seasonal, for example) (Baldwin and Dunkerton, 2001). It has been suggested that ozone depletion over Antarctica can influence the variability of the so-called Southern Annular Mode, and it has been shown that changes in PWD in the stratosphere can change the variability associated with the Northern Annular Mode. Although the component of the ACC response that projects onto the annular modes may have a relatively large amplitude, its detection may be challenging because, by definition, the annular modes are patterns of internal variability. A major challenge for SPARC will be to understand the nature of this stratosphere-troposphere coupling, as well as to determine the spatial (in particular, vertical) resolution needed in climate models to capture the relevant dynamical processes.

In the presentation to the JSC on this topic, we noted that the three main SPARC themes are all relevant to climate change. The SPARC community makes important and ongoing contributions to ACC research and assessments. CCMVal provides key input to the UNEP/WMO Ozone Assessments by assembling, archiving and analysing reference CCM simulations. Investigating dynamical features of ACC is a key aspect of the stratosphere-tropophere dynamical coupling theme. Examples include studies of changes in planetary wave activity and the Brewer-Dobson circulation associated with ACC and the influence of stratospheric changes on patterns of variability such as the annular modes. The theme on detection and attribution of stratospheric changes has ongoing activities assessing and updating of stratospheric temperature trends, assessing the role of transport in modelling chemistry and ozone recovery and characterising variability in the stratosphere. These activities will be enhanced in the context of the dynamical modelling strategy that was proposed in the SPARC presentation (see the accompanying report on the JSC meeting in this newsletter).

Other issues raised in the ACC discussion

Although not all have explicit programme elements for this purpose, all of the core WCRP projects have ongoing activities that are relevant to understanding and predicting ACC. Cross-cutting activities, such as evaluating and comparing the performance of climate models used for assessments (such as for the IPCC AR4) require collaboration between the core projects, working groups, and panels. Presentations at the JSC meeting summarised relevant current and anticipated WCRP activities. Some notable issues that were discussed include investigating the role of land use changes in ACC and assessing key uncertainties in climate prediction.

Profound changes in land use have accompanied industrialization in the past two centuries. A well known example involves the development and spread of agricultural activities. Equally important developments include the extensive changes in river flow regimes that have accompanied these activities, as well as the accelerating growth of large urban centres (especially in Asia). There is increasing evidence, from both modelling and observational efforts, that such changes have important effects on the climate system. The GEWEX presentation summarised current and planned activities that address evaluating the role of land-use changes in climate change. These are being carried out within the GEWEX Global Land-Atmosphere System Study (GLASS).

Evaluating uncertainties in climate prediction is a key part of the CLIVAR focus on ACC. In collaboration with WGCM, CLIVAR has promoted global and regional analyses of IPCC AR4 simulations and is undertaking an assessment of the seasonal prediction capabilities of IPCC class models. Principle uncertainties in predicting ACC on sub-century time scales are associated with sub-grid parameterizations, which has consequently motivated a more probabilistic approach. The prediction of PDFs of ACC, though potentially valuable, is a nascent approach which requires progress on resolving key uncertainties. Stochastic-dynamical approaches to parameterization are in early stages of development, and characterising model uncertainties is a key requirement for evaluating uncertainties in the prediction of PDFs of ACC.

Evaluating, characterising and reducing uncertainty in modelling of the stratosphere, and evaluating the importance of resolving the stratosphere in prediction are central to the SPARC dynamical modelling strategy. The COPES seamless prediction perspective provides a valuable context to move forward on the issue of reducing uncertainty in ACC prediction. Evaluating modelling errors in predictions ranging from short-range forecasts in the numerical weather prediction context to seasonal and longer time scales, in combination with utilising the tools and outputs of data assimilation systems, has the potential to characterise and reduce uncertainties in modelling of key processes.

JSC Actions on ACC

The JSC, while noting the great deal of effort going on in various ACC activities of WCRP, identified as a key issue the need for the WCRP to raise its ACC visibility to a higher profile. A roadmap for ACC Activity is to be developed by a JSC task team, recognizing the existing work (with contributions by the projects, groups, task teams *etc.*), and proposing how the WCRP can deliver on its objective to determine the effect of human activities on climate. A first draft will be available for the WCRP Officers, Chairs, and Directors meeting which will be held in Beijing immediately preceding the ESSP Open Science Conference in November, 2006.

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Getting to GRIPS with Climate-Middle Atmosphere Model Validation

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For about a decade, the GCM-Reality Intercomparison Project for SPARC (GRIPS) was the central activity for SPARC's model evaluation. This article summarises, from a personal perspective, how GRIPS has contributed to SPARC's goals and how the legacy of GRIPS should impact SPARC's future modelling activities. I dedicate the article to my GRIPS co-coordinator, **Kunihiko Kodera**, in honour of his recent retirement from the Meteorological Research Institute in Tsukuba, Japan.

GRIPS focused on the dynamical and radiative aspects of stratospheric climate and its interactions with the troposphere. Participation was limited to models that could loosely be termed "Climate-Middle Atmosphere Models" which was defined on the basis of three features: the upper boundary was at or above the stratopause, the water cycle was included, and the model used a full radiation code. These conditions precluded a number of "approximated" models that were commonly used in the early 1990s, because the central goals of SPARC concern the interactions with climate. Many of the models that participated in GRIPS were vertical extensions of models that were being used for climate studies, with an upper boundary in the low to middle stratosphere.

Evaluation and validation of models generally require some "rules" to ensure that similar scenarios are being compared. An early example for GRIPS was the Atmospheric Model Intercomparison Project (AMIP), led by **Larry Gates** of the Program for Climate Model Diagnosis and Intercomparison (PCMDI) at the Lawrence Livermore National Laboratory in the USA. Larry generously devoted time and resources to assisting GRIPS, including attending workshops and inviting me to PCMDI to interact with his colleagues. AMIP's goal was to assess the performance of tropospheric climate models run under conditions with identical surface forcing (sea-surface temperatures and ice distributions); a list of mandatory output was also specified. To meet these demands, PCMDI developed an infrastructure to collect, quality-control, and analyse the AMIP output.

While the AMIP model clearly met many requirements to ensure successful model evaluation, it soon became apparent that it was not appropriate for GRIPS. A main issue was cost. An infrastructure equivalent to that at PCMDI requires substantial support that GRIPS could not offer; furthermore, each of the participating model groups did not have the resources for additional simulations, so it was not possible to meet the "standards" necessary for an AMIP-like structure. Instead, it was agreed that we would collect and analyse output from simulations that were being performed in the various institutions. No data formats were imposed: data were simply collected along with reader routines and processed into reasonably compatible formats. After a few iterations, a suite of monthly mean output files from 13 models was collected and key results were published in Pawson *et al*. (2000). That paper lists a set of "Phase 1" tasks for GRIPS, one of which was "documentation" – one of the lessons learned from AMIP was the need to rigorously document the models that were analysed, a lesson that was adhered to by more than half of the GRIPS models!

The absence of substantial funding for GRIPS was an impediment. A small group of European modellers did have modest support through "EuroGRIPS" for some time, while others were able to include specific GRIPS tasks in their research budgets. However, there was never a fully funded project scientist or data manager for GRIPS, which slowed down progress for an intended "second paper" which would look more at forcing mechanisms. This had an impact on my ability to contribute, because after I moved from a rather flexible university environment to a more project-oriented institution, I was not able to devote as much time to GRIPS; my other responsibilities of journal editorship and increasing family size also had a large impact there.

Without the baseline funding, progress came down to individual scientists having the time and motivation to carry out specific tasks. Excluding documentation and model climate, seven "Phase 1" tasks are listed in the BAMS paper. These tasks all involve analysis of results from existing model runs, and through substantial efforts of numerous scientists, five of these seven tasks were brought to closure, with either brief reports or peer-reviewed papers. The studies of Koshyk *et al*. (1998) and Horinouchi *et al*. (2003) both arose from GRIPS and share a story of enthusiasm and initiative. Each involved a young scientist attending a workshop with an idea about processes that they wished to investigate: **John Koshyk** was interested in spatial wavenumber spectra and **Takeshi Horinouchi** wanted to examine relationships between convective parameterizations and the excitation of tropical waves. Having presented their ideas and defined the data they needed to analyse, they next coordinated with the individual modelling teams to collect the data and complete the analyses, leading to eventual publication of their work. The efforts of John and Takeshi are just two examples of GRIPS projects that succeeded, and I stress that many other scientists (too many to name) made similar contributions.

The "second" and "third" phases of GRIPS built on from the results of the first phase, requiring the groups to perform additional model runs. "Phase 2" actvities were designed to investigate sensitivities to different parameterizations, especially radiation and gravity-wave drag. No publications have directly arisen from these tasks, but much interesting discussion and understanding was gained. The radiation comparison was motivated by the cold bias in the models, as discussed in the BAMS paper. Detailed comparisons were presented at conferences (by **Ulrike Langematz** at IUGG in 1996 and by **Piers Forster** at the SPARC 2004 Assembly), highlighting several uncertainties in the radiation codes.

Subsequent studies by various model groups have shown that the cold stratospheric bias is smaller when the radiation code is better.

The third phase of GRIPS involved model runs intended to isolate how various forcing mechanisms might impact the stratosphere, including their variability and their effect on climate. These included solar forcing, volcanic aerosols and ozone change (specified in the models) between 1980 and 2000. A paper by Matthes *et al*. (2004) attempted to untangle the many influences that interact to yield a solar signal in the atmospheric structure. Such signals are difficult to extract in the stratospheric system where the interannual variations are large; this point arose many times over the course of GRIPS, including the need for an adequately long record to determine a stable climatology, which also meant that attempts to isolate potential stratospheric impacts on the troposphere were not fruitful, at least in the first few years of the project.

Key to the evolution of GRIPS was the annual workshop. These served as the venues to meet and present results and ideas that were fully discussed. It was not necessary to bring a polished, completed piece of work that could be presented at a conference. We encouraged people to show more of the basic progress and emphasised discussion and interaction, so that we could learn from each other's progress or misfortune! This developed a cordial atmosphere of collaboration and generated many friendships among the participants. The workshops were generously hosted at a range of institutions; I single out the Canadian Middle Atmospheric Modelling group for special acknowledgement, since they hosted the first and last GRIPS workshops (in Victoria and Toronto). I also extend special thanks to **Elisa Manzini** who organized two workshops, the second in Bologna just a few years after I had to withdraw from the first one she had hosted in Hamburg. My non-attendance of the Hamburg workshop gives **Kunihiko Kodera** the honour of being the only person to have attended all ten GRIPS meetings!

Over the lifetime of the GRIPS initiative, many things changed. Several models were retired and groups combined forces to produce newer models, leading to better agreements among the later models than in the BAMS paper. I like to think that the GRIPS comparisons impacted these developments. Increases in computational power have been substantial in the past decade and this has facilitated longer model runs, which are essential if we are to fully characterise the atmospheric variability and separate signals (if any) of causal forcing mechanisms from the random variability of the circulation. However, by far the biggest advance is the ability to include complete chemistry codes in the climate models, in order to perform multi-decadal predictions to investigate the evolution of the ozone layer as chlorine levels decrease and greenhouse gases increase. In later years, more and more discussion at GRIPS workshops was devoted to coupled chemistry-climate modelling. Initially the work was exploratory and performed by relatively few groups. However, around the time of the 2002 WMO-UNEP ozone assessment several groups were able to produce long model runs that examined ozone in the 21st century (see Austin *et al*., 2002). Given the environmental importance of the ozone layer, it became imperative that some international coordination was needed to evaluate the models and ensure that future predictions are performed with similar conditions. This could have been possible through a "Phase 4" of GRIPS, an issue that was discussed at GRIPS meetings and SPARC SSG meetings, but because of lack or resources and time this was not feasible. SPARC opted to take a more radical approach and encouraged the formation of the Chemistry-Climate Model Validation (CCMVal) activity, which is much broader in scope than GRIPS. CCMVal faces many challenges, but there is precedent in SPARC's past success. I am very optimistic that CCMVal will continue with evaluation of chemistry-climate models and will build on the successes of GRIPS and other similar activities. When I reflect on how GRIPS was successful, I come back to a couple of the points mentioned in this short article. The key is in the teamwork. The coordinators can achieve a lot, but not without support and enthusiasm from the community. I especially encourage any younger scientists to bring your ideas to CCMVal workshops: present your concepts and encourage some of the model groups to share their data with you. Starting small (one or two models) can provide the basis for validating the usefulness of an idea, to the point that others may want to join in. Interactions like this can lead to fascinating insights and can evolve into interesting opportunities for collaboration – to me this remains a great motivation and source of optimism for the success of CCMVal and other SPARC-WCRP modelling activities.

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I am grateful to many people who have supported the GRIPS effort. At the Free University of Berlin, **Karin Labitzke** introduced me to the SPARC community and encouraged me to take on a role in their modelling activities. **Ricky Rood** and **Michele Rienecker** continued to support my involvement after I moved to NASA GSFC. **Marie-Lise Chanin** and **Marvin Geller** were inspiring leaders of SPARC for several years, a tradition continued by the present chairs, **Ravi and Alan.** The WCRP supported GRIPS in many ways, often providing partial support for the workshops: without wishing to underestimate the wide-ranging contributions of WCRP staff, it is justifiable to single out **Roger Newson** for special thanks.

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Dynamics of the stratospheric circulation response to climate change

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Recently many modelling groups around the world have employed chemistry climate models (CCMs) to predict the future stratospheric dynamical and chemical environment. These results are providing essential input into the WMO/UNEP Ozone Assessments (Eyring *et al*., 2005). Despite the abundance of simulations, however, many dynamical issues remain unresolved. In particular, it is not clear how planetary waves and their associated forcing of the Brewer-Dobson circulation will respond to climate change. While the SPARC CCMVal activity will assess the dynamical capability of CCMs with key validation diagnostics, understanding the underlying causes of the models' dynamical behaviour is likely to require more detailed investigation. Accordingly, we propose to establish a SPARC subproject within the context of SPARC's "stratosphere-troposphere dynamical coupling" theme, which will address some of the outstanding dynamical issues in a systematic way. With the help of a variety of dynamical tools and a hierarchy of models, we aim to identify robust dynamical mechanisms that may be expected to be operative in the real atmosphere and in CCMs.

The dynamical issues are relevant to SPARC as a whole because of the connection of planetary wave forcing to the Brewer-Dobson circulation (BDC). The BDC is a meridional circulation that consists of rising motion in the tropics, a poleward drift towards the winter pole, and downwelling in the extratropics. This circulation is induced by the breaking and dissipation of waves that emanate from the troposphere. The BDC is usually characterised by either the diabatic or the residual circulation, and is essentially the stratospheric *mass* circulation. Its strength and structure affects stratospheric temperatures (and hence winds) through dynamical heating or cooling. While the BDC also has obvious implications for transport, it is to be distinguished from the stratospheric *transport* circulation, which determines the age of air. The latter is determined not only by the BDC, but also by other processes such as horizontal mixing, whose strength is different for different chemical species and thus cannot be characterised in a simple manner.

Changes in the BDC and related changes in the strength of the stratospheric polar vortex have important effects on atmospheric chemistry through their influence on wintertime polar temperatures, which in turn influence, for example, rates of chemical destruction of ozone; through the transport of ozone and other chemical tracers and through their control of the strength of stratosphere-troposphere exchange. To the extent that the strength of the stratospheric polar vortex influences the wintertime circulation in the troposphere — and there is increasing evidence for such an influence — trends in polar vortex strength can also result in regional and hemispheric trends in tropospheric climate, such as those associated with trends in the strength of the Arctic and Antarctic Oscillation indices. A stronger BDC also increases the transport of stratospheric ozone to the troposphere, possibly leading to higher tropospheric ozone concentrations (Collins *et al*., 2003).

Recent studies

Butchart *et al*. (2006) have compared the effects of climate change on the tropical upwelling in a number of middle atmosphere general circulation models (specifically, those involved in the GRIPS project [GCM Reality Intercomparison Project for SPARC]). They find that all models show an increase in tropical upwelling in response to climate change (either from greenhouse-gas increases or the combined effects of greenhouse-gas increases and ozone depletion). The multi-model mean trend is about 2% per decade, but the trend varies considerably between models. More than half of the trend can be attributed to changes in the explicitly resolved wave drag.

In contrast, the sign of the response to climate change of the polar downwelling differs from model to model. Austin *et al*. (2003) have compared the results of several CCMs for the Arctic. They considered the wintertime meridional heat flux in the mid-latitude lower stratosphere, a quantity that provides a measure of the wave forcing from the troposphere and is highly correlated to polar downwelling (Newman and Nash, 2000). Most models showed a gradual long-term reduction of the lower stratospheric heat fluxes in the past two decades (consistent with observations, albeit weaker), but for the future, the results are more mixed; most models continue to show a slight decrease (associated with less polar downwelling), but at least one model (E39/C) shows a significant upward trend (**see Figure 1**).

Focusing on single models, Sigmond *et al*. (2004) and Fomichev *et al*. (2006) have reported on the simulated effects of CO_2 increases on the BDC, distinguishing the direct stratospheric radiative response from the response induced by changes in tropospheric climate. The former study finds a stronger descending branch of the BDC in the doubled CO_2 climate. This is associated with more wave activity just above the tropopause (which is then dissipated in the lower stratosphere), whereas they find decreased wave activity in the troposphere. Two plausible causes of the increased lower stratospheric wave activity are: 1) there is more wave production in the tropopause region or 2) the tropopause region is more transparent to the propagation of tropospheric wave activity.

In summary, the expected response of the BDC to climate change remains unresolved. There are some hints of robust

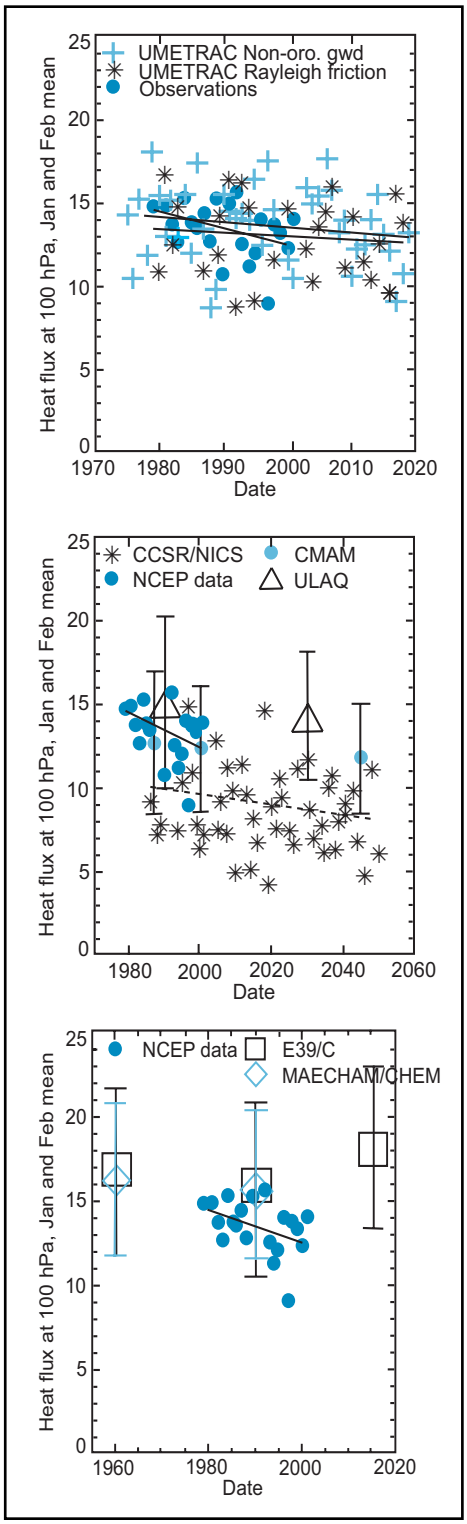

Figure 1: Scatter diagrams of the winter-time mid-latitude heat flux at 100 hPa (a proxy for the polar downwelling) plotted against year for NCEP data (black line in all panels), and for a set of CCMs. Upper panel: transient runs with a model with Rayleigh friction or non-orographic gravity wave drag. Middle panel: transient runs with three different CCMs. Lower panel: time-slice runs with two different CCMs. (From Austin et al. 2003)

responses in the tropics, but the response of the planetary waves and of the descending branch of the BDC are still uncertain.

Outstanding questions

In order to have confidence in model projections of changes in the BDC it is necessary to understand the dynamical processes responsible for these changes. Some outstanding dynamical questions we have identified include:

1) How does stratospheric wave activity respond to climate change, and how does this affect tropical upwelling and polar downwelling? Do the wave activity changes arise primarily from changes in the wave flux from the troposphere, or from changes to the zonal mean winds and temperatures that influence the environment for planetary wave propagation (as indicated by changes to refractive indices or to reflective surfaces, see Harnik and Lindzen, 2001)?

2) Is there a contradiction between the apparent consensus between different models regarding the response of tropical upwelling to climate change (Butchart *et al.*, 2006), and the more mixed results in the Arctic (*e.g.*, Austin *et al.*, 2003)? Can this be explained by changes in the structure of the BDC, in which case the tropical upwelling and polar downwelling may change independently of each other?

3) Which part of the BDC response to climate change is attributable to greenhouse warming/cooling and which to ozone depletion? This might be a surprisingly difficult question to answer from observations as evidence accumulates that both greenhouse warming and ozone depletion cause similar annular-mode type responses in the zonal-mean circulation of the stratosphere and troposphere.

4) To what extent are the models sensitive to their treatment of unresolved (*e.g.* gravity) waves and other dissipative processes? This may be a question of particular importance in understanding the climate-change response of the Southern Hemisphere, in which gravity wave drag on the vortex is presumably very important. Shaw and Shepherd (2006) have recently argued that the gravity wave drag response to a climate perturbation should be much more robust if angular momentum is conserved in the implementation of the gravity wave drag parameterization something that is generally not done in practice). Additionally, how are the simulated BDC and its sensitivity to climate change affected by model resolution and lid height?

Proposed SPARC subproject

Given these and other questions, we propose to establish a SPARC subproject to understand the dynamics of climate change in the stratosphere, with a focus on the changes in the wave driven BDC. The idea is to analyse a wide range of models with a well resolved stratospheric component, which might or might not include interactive chemistry. We propose to start with existing integrations, and to develop tools that could be quickly applied to new simulations as they become available, for example through the IPCC AR4 archive and CCMVal. Thus, this activity will help develop and refine robust dynamical diagnostics that can be subsequently included within CCMVal. We divide the activities of this subproject into the following tasks:

Task 1: Baseline diagnosis of changes to the stratospheric zonal-mean circulation and to stationary-wave structures, and resulting changes to wave driving and to the BDC

The first step is to analyse the response of basic quantities to climate change in different models, using existing simulations from GCMs and CCMs. These quantities include temperature, wind, geopotential height, the residual stream function, wave activity and wave driving. It will also include an analysis of the eddy amplitude and the phase structure for individual planetary-scale waves (wave number 1-4) and the response of these waves to climate perturbations. Our initial focus will be upon the stationary wave field of Northern Hemisphere winter. The key question here is if we can identify a set of common responses among the models, and common dynamical mechanisms acting in them.

Task 2: Analysis of stratospheric wave responses using linear models

This step will use stationary wave models to analyse the response of the stratospheric

portion of the stationary wave field to climate perturbations. Following the method of Ting *et al*. (1994), we will analyse the separate effects of changes to the different zonally asymmetric forcings (diabatic heating, the interaction with the transient eddies, and the interaction of the stationary eddies with the basic state). Finally, there is limited evidence (see Joseph *et al*., 2004) that, in the troposphere, the changes to the zonal mean circulation (as opposed to changes in the zonally asymmetric forcings) heavily influence the stationary wave response to climate perturbations. An open question is if this would also be the case for the stratosphere.

Task 3: Analysis of changes to the stratospheric wave environment

As a complement to the stationary wave analysis of Task 2, we propose to carry out an analysis of changes to the stratospheric environment as a medium for wave propagation. This task would involve calculation of different indices of refraction and reflective surfaces, largely based on the climatological field output from the models (*e.g*., Perlwitz and Harnik, 2003). These linear modelling approaches would complement the stratospheric stationary wave model described in Task 2.

Task 4: Response of simplified GCMs to stratospheric and tropospheric eddy forcing changes

The idea of this task is to use simplified atmospheric GCMs to study how the zonal-mean extratropical circulation responds to imposed eddy forcing derived from climate change runs (*e.g*., Kushner and Polvani, 2004). One such approach, following Hall *et al*. (2001), is to tune the diabatic heating in a dry 'dynamical core' GCM to fit the circulation climatologies of the various models.

Conclusion

Despite the abundance of simulations of the future stratospheric climate, many dynamical issues remain unresolved. In particular, it is not clear how the wave forcing will respond to climate change. General circulation models predict an increase of the tropical upwelling in response to climate change, but the results in the extratropics are less clear. By clarifying the dynamical mechanisms of the responses we hope to be able to place more confidence in model projected changes in the strength of the BDC and its implications for chemical and physical climate.

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The routes for organics oxidation in the atmosphere and its implications to the atmosphere

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Despite a large number of process studies over the past decades, which have improved our understanding of ozone chemistry, many processes fundamental to atmospheric chemistry are still poorly understood or need to be described in greater detail, in order to furnish a scientifically sound basis for the forging of better assessment tools in terms of chemical and physico-chemical process modules which can be included in numerical models of our atmospheric environment. A timely example is the chemical, physico-chemical, morphological, optical and toxicological characterization of aerosol particles. This information is needed to assess their impact on air quality (*e.g.* visibility reduction), on climate, and on human health. Another example is the development of structure-activity-relationships for various classes of new volatile or semi-volatile chemicals, which are needed to assess their tropospheric lifetimes, and the number, structures, and properties of their degradation products, their impact on ecosystems, and their potential to contribute to new particle formation.

In addition, hydrocarbons play many important roles in the atmosphere:
(1) They provide the "fuel" for the production of photochemical ozone in the troposphere,
(2) They produce chemicals that lead to the formation of new aerosol particles,
(3) They condense on existing aerosols and alter their properties with respect to their ability to take up water, form ice and cloud condensation nuclei, and the optical properties of the particles, and
(4) They are the carriers of key minor, but important trace species *(e.g.,* chlorine, bromine, and sulfur) that when released *via* the oxidation of the organics, play key roles themselves (*e.g.,* release of chlorine and bromine in methyl chloride and bromide in the stratosphere leads to ozone destruction in this region).

Because of these reasons, understanding the role of organics is key for quantifying the role of organics in many environmental areas, including the role of organic containing aerosols in climate forcing and regional air quality and the production of tropospheric ozone, which again is a key climate and air quality agent.

To trigger actions on such questions, a workshop, was initiated by **C. George** and **Y. Rudich**, and was jointly organised with **R.A. Cox, J. Hjorth, M. Pilling, A.R. Ravishankara** and **P. Wiesen**. This meeting emphasised the routes for organics oxidation in the atmosphere, aerosol formation and properties, and the implications to the atmosphere (both in atmospheric chemistry and air pollution), and aimed at bringing together scientists to exchange the latest data and ideas through intensive discussions. The meeting was broken into sessions aimed at anwering a few important questions

- Can we predict the atmospheric fate of complex (biogenic and multifunctional) organic compounds ?
- How well is low temperature gas phase organic chemistry understood ?
- The role of organics in new particle formation, growth of particles. How well do we understand aerosol abundance, properties and mass balance ?
- High molecular weight species in aerosols: What is the role of *in situ* polymerization versus other mechanisms and what is its implication to the atmosphere ?
- How can organic compounds affect atmospheric ice nucleation processes and the properties of cirrus clouds ?
- How well is the reactive uptake of (organics) gases by an organic and/or ice surface quantified/understood ?

Each session was aimed at addressing the above questions and was introduced by several invited speakers who were expected to cover some overall general issues as well as some specific questions. Discussion leaders had the task of motivating the discussion and preparing informal late evening discussions. Several poster sessions helped to supplement the material presented in the oral sessions.

Session 1: Can we predict the atmospheric fate of complex organic compounds?

The introduction for this section was given by **N. Donahue** who showed that currently, a general scheme exists, at least in principle, for OH radicals initiating the removal of an organic emitted at the surface that gets into the atmosphere (referred to as volatile organics, VOCs) and converting them into various chemicals and eventually carbon dioxide. However, this mechanism becomes progressively less accurate as the size and complexity of the organic compound increase, *i.e.,* with increasing carbon number and functional groups (with oxygen), and especially at low abundances of NO_x. Our knowledge of the oxidation mechanism is summarised in the Master Chemical Mechanism (MCM), an extensive sequence of reactions generated

from detailed chemical knowledge. This mechanism is a crucial link between laboratory derived kinetics data and the reduced mechanisms used in large-scale models.

Currently, chemistry taking place when the abundance of NO_x is low is still poorly understood for major emissions such as isoprene. There is growing evidence that a reaction sequence can become substantially more complicated as the VOCs become more complicated. Examples of the complexity include production of unsaturated cyclic compounds (dihydrofurans) from 1,4 hydroxy carbonyls, which are in turn oxidation products of larger VOCs, as well as more prevelant photolysis and subsequent production of Secondary Organic Aerosols (so-called "SOA") from ortho aromatic aldehydes. Mechanisms that capture these complications are needed to predict ambient abundances of stable and reactive species, to predict generation and removal of ozone, to predict organic aerosol production, and understanding aging of organic aerosol particles and organics in condensed phases. We are in good shape in some cases, such as ozone and radical production and loss at high NO_x, but we are still far from answering the guiding question affirmatively in most cases.

In the introduction to this workshop, **A. Ravishankara** stated that 'complexity of organics has perplexed us'; the initial session served to support his contention. Increasing size and substitution in organic compounds clearly leads to increasing complexity in oxidation mechanisms. The current simple schemes for the possible pathways in the oxidation of large (C5 and larger) organic compounds is likely to be inaccurate. One reason is the number of possible reaction pathways that are open to a large molecule in the atmosphere, including radical attack at numerous sites, photolysis rates, and occurrence of isomerization reactions. A second reason is that the atmospheric fate of these large compounds includes a succession of individual steps, progressing from one compound to the next through an unknown number of steps before removal of the species from the atmosphere *via* deposition. At a minimum, the possible number of "generations" (*i.e.*, individual steps in converting the molecule to CO_2) is the number of carbon atoms in the precursor. The consensus of the meeting was that we need more than ever to integrate a range of approaches to constrain this problem, and that the necessary constraint is the actual consequence of these compounds in the atmosphere.

In the first two talks of this session, the speakers described encouraging recent advances in the availability of better analytical methods in recent years. In the first talk, **R. Atkinson** showed that hydroxyl-substituted carbonyls (1,4 hydroxycarbonyls) can cyclize and eliminate water to form dihydrofurans (unsaturated cyclic organics with a single oxygen in a 5-member ring). Not only is this an example of the rich chemistry that can emerge with size and substitution, but also evidence of what can happen in the atmosphere. The dehydration step requires relatively low relative humidity (< 35%), but the precursor hydrocarbons are found in urban plumes, many of which advect over arid regions (for example, Los Angeles) with low humidity. Unsaturated dihydrofurans formed in these processes can react with ozone and produce low vapour pressure products that form secondary organic aerosol (SOA).

A second example presented by R. Atkinson concerned the oxidation of aromatic compounds by OH radicals, which is also of great importance to the chemistry of urban plumes. Here, recent analytical advances have enabled detection of dicarbonyls, which again appear to be important contributors to SOA formation. The significance of SOA, in turn, is two-fold. It may contribute substantially to the total fine-particle mass burden, which is a major health concern, and organic particles or particles containing organics, especially those organic compounds that are oxidized and relatively polar (or 'aged'), also participate in the hydrological cycle as cloud condensation nuclei and thus play a role in the indirect climate effect.

In a second talk, **W. Mellouki** presented recent experimental data describing the ozonolysis of oxygenated alkenes as well as the photolysis of aromatic aldehydes. The first subject ties in directly with the discussion of dihydrofuran ozonolysis as another example of 'later-generation' oxidation steps of potential importance to the atmosphere. This is also true of the discussion of photolysis, which raises the last of the important factors governing the fate of organics. Two fascinating aspects of the photolysis were discussed: (i) the position of substituents in dialkyl benzenes has a profound influence on their photolysis rate, and (ii) the photolysis products include compounds formed by cyclization. Once again, substantial SOA formation is involved.

The frequent evidence for SOA formation and the enormous richness of the chemical palette associated with SOA formation points again toward the need for simplifying structures articulated in A. Ravishankara's overview. In this case, the critical issue is the distribution of vapour pressures in oxidation sequences, including any simplifying generalizations we can make to describe the evolution of vapour pressures through multiple generations of oxidation. There are two general approaches to MCM development: i) a 'comprehensive' method (close to the complete enumeration described above) involving extensive estimation of unknown rate constants through automated semi-empirical methods, and ii) an 'explicit' approach focusing on measured rate constants with a certain degree of extrapolation to important unmeasured reactions by hand. **M. Pilling** focused on the second approach, showing that there is a clear benefit to having a mechanism that aims to describe 'complete' chemistry as we know it (in the gas phase), but that the emerging complexity described here presents an enormous challenge, requiring a collective, community-wide response. A protocol for updating the mechanism as new results become available is critically needed – updating 50,000 reactions by hand annually would require considering 1000 reactions per week.

One specific example discussed in detail was the photo-oxidation of toluene. The current representation in the MCM fails to reproduce toluene oxidation data from chamber experiments, mostly because the radical production rates in the pure toluene oxidation appear to be significantly greater in reality than in the mechanism, in a way that does not generate ozone efficiently. On one hand this shows how the MCM can be used to close the loop between experiment and theory (presuming that the missing radical source can be identified), but it raises the question of whether the 'missing' radical source is relevant to the atmosphere. Ultimately, this is another example of 'perplexing complexity' that may be best answered through careful field stu-

dies, rather than exclusively in controlled chamber and elementary kinetics experiments. The ultimate conclusion of this session was that while we have made substantial inroads into the well-known problem areas regarding organic oxidation, we are still far from answering 'yes' to the introductory question. As much as we lack a detailed answer, we may still lack the correct paradigm for the answer. This is a greater concern.

Session 2: How well is low temperature gas phase organic chemistry understood?

C. Nielsen started this discussion item followed by a presentation by **P. Shepson** on photochemistry of organic compounds in ice. Numerous observations in polar regions have shown that photochemical processing in the top 10 cm of snow releases substantial amounts of oxygenates to atmosphere. This release has an important impact on the local chemistry. Snow is rich with organic reactants - humic material is often the dominant organic substrate - and H_2O_2 is the dominant precursor to condensed phase OH. Reactant concentrations are often very high, and pH's are very low. Organic photochemistry and photooxidation is not well characterised in snow and ice, and much field work remains to fill the gaps in our understanding of the mechanisms. Measurements of the isotopic composition may give useful information. There is clearly a need for laboratory data on absorption spectra and quantum yields for photolytical processes in ice.

In addition, the chemistry of the upper troposphere (UT) is not fully understood. **G. Lebras** presented experimental studies of oxidation of oxygenated VOCs at the low temperatures experienced in the UTLS region, showing that measured HO_x concentrations (OH and HO_2) are 2-4 times higher than modelled when considering only ozone photolysis and methane oxidation. In addition, substantial amounts of oxygenated VOC's have been observed in the UT, and their photo-oxidation has been suggested as the missing source of HO_x in models. The consequences of enhanced HO_x concentrations are: (i) increased rates of oxidation processes and net ozone production for given NO_x levels, (ii) increased sensitivity of ozone production to increasing emissions of NO_x from aircraft emissions and from the polluted boundary layer. The available experimental low temperature studies (down to 200 K) show that the temperature dependence of the rate constants is not captured by the Arrhenius law. Also product yields, absorption cross sections and quantum yields for photolysis show strong temperature dependencies. There is a clear need for more experimental data at low temperatures.

L. Vereecken presented computational difficulties encountered in *a priori* predictions of OH-initiated oxidation of oxygenates at low temperatures. While theoretical calculations have been successful in many areas, *e.g.* for Structure Activity Relationships (SARs) for alkoxy radical reactions, in the case of the OH-initiated oxidation of oxygenates such calculations are notoriously problematic due to H-bonding, adduct formation, small barriers and tunnelling effects. Experimental results for the OH reactions of acetone, hydroxyacetone, formaldehyde and acetaldehyde, provide a general idea of the potential energy surfaces and trends to expect, and where the computational difficulties rest. For apparently simple reactions such as acetic acid+OH, formic acid+OH, and glycolaldehyde+OH, the reported theoretical work is often inconsistent with the experimental observations. More low temperature experimental data are needed to resolve many of these inconsistencies and to improve our understanding of such reactions. Laboratory studies of kinetic isotope effects may also be of great value in this respect.

Session 3: Role of organics in new particle formation, growth of particles. How well do we understand aerosol abundance, properties, mass balance?

Increasing attention is being focused on atmospheric aerosols and on their influence on air quality, climate and atmospheric chemistry. One of the major parameters which determines the impact of aerosols on health, radiation and cloud microphysics is particle size. As shown by **T. Hoffmann**, several processes determine the aerosol size distribution, including new particle formation by gas-to-particle conversion, growth due to condensation and coagulation, the emissions of primary particles and removal by dry or wet deposition. New particle formation is probably the least understood of these steps, although in recent years, the formation and growth of nanometer-scale particles have been observed *in situ* in many different atmospheric environments, including urban plumes, clean Arctic air, the continental boundary layer and coastal environments.

Within the last two decades, the discussion about the chemical nature of the nucleating species has primarily focused on sulphuric acid, since sulphate is an important component of the nucleation mode aerosol. Meanwhile, it is generally recognised that binary nucleation of sulphuric acid-water is not efficient enough to explain atmospheric new particle formation. Recent progress has been made in assessing the importance of ternary water-sulphuric acid-ammonia nucleation, ion-induced nucleation as well as nucleation involving iodide species. However, still an open question is the topic of this session – the role of organic compounds in new particle formation.

Since a direct chemical analysis of the newly formed atmospheric particles is not possible, the research on this topic requires an interdisciplinary approach involving laboratory experiments, continuous field observations, new theories and dynamic models to determine which mechanisms dominate in different environments. This was also obvious within the session, where the first two presentations, by **K. Carslaw** and **M. Kanakidou**, focused on modelling aspects of particle formation from the oxidation of VOCs and the last contribution, by **A. Laaksonen**, concentrated on the interpretation of laboratory and field work. The presentations and the intensive discussions during the session showed that organic compounds seem to be involved in most types of nucleation events observed in field studies, except perhaps in very polluted regions. It is still an open question if organics are just involved in the growth of TSCs or if they can also influence the early steps of cluster formation.

Due to some advances in some parametrizations, new particle formation schemes can be incorporated into global models and they can successfully reproduce surface observations of particle number quite well. The relative importance of given organics is strongly influenced by the local environment. When new particle formation happens in boreal environments,

organics dominate the particle growth and monoterpene oxidation products appear to be the most important condensing species. When new particle formation takes place in polluted continental environments, there exist strong indications that organic compounds are often involved in the nucleation events; however, possibly their role varies with season. Finally, the contribution of new particle formation and growth to CCN is potentially large, but currently uncertain.

Session 4: High molecular weight species in aerosols: What is the role of in situ polymerization versus other mechanisms and what is its implication to the atmosphere?

It is increasingly apparent that a significant fraction of submicron aerosol is organic in nature and that much of this material is composed of high molecular weight material. However, very little is known about the composition, and physical and chemical properties of the high molecular weight material. Since this fraction forms such a large component of submicron aerosol, its atmospheric lifecycle needs to be understood to predict its impact on cloud activation, water equilibrium and hence radiative impact, and its effect on heterogeneous chemistry. This session, introduced by **H. Coe**, summarised what is known about the chemistry of this fraction, and what is known about the formation pathways and fluxes from the gas phase.

U. Baltensperger gave an overview of recent results on the formation of SOA in chamber studies. In every organic system studied to date new particles were nucleated in the absence of seed aerosols and rapidly grew in size showing that volatility decreases over time, with evidence of polymeric, or more precisely oligomeric, development. In addition to the formation of oligomers, there was clear evidence for changes in chemical functionality, in particular FTIR analyses showed the development of organic acid groups. A wide range of new techniques and instruments have recently been used to measure the chamber aerosol, however, at present no instrument currently provides a complete picture of the oligomeric material, its molecular weight and chemical composition and it remains unlikely that a single method will achieve this.

A question remains as to whether isoprene is an important precursor of oligomeric organic aerosol. Evidence was presented that isoprene could be an important precursor, though the conditions under which these experiments were carried out may not be relevant to the atmosphere, since either high precursor concentrations or high oxidant concentrations have been used. Ambient samples taken in Zurich during the summertime show similar chemical signatures to those observed in smog chamber SOA, but wintertime aerosol show different characteristics, being much larger molecular weight material.

G. Kiss illustrated the importance of establishing the molecular weight of the oligomeric fraction in cloud droplet activation through its effect on the hygroscopicity. It was shown that typical molecular weights peak at around 250 Da but cover a range from 100 to 700 Da. Aqueous phase reactions can also produce high molecular weight compounds that are coloured and may significantly affect the absorption properties of the aerosol. These reactions require further study. It is clear that the "humic-like" substances are far less hygroscopic than inorganic ions, and in mixed component aerosol the growth is almost entirely due to the inorganic fraction. During cloud activation the organic fraction may affect the surface tension and subsequent growth or evaporation processes. The organic fraction significantly adds to the mass of any CCN and therefore increases the likelihood that a particle activates.

H. Herrmann presented results on the formation of SOA in chamber studies where acid seed aerosols are present. Product studies revealed that particle phase heterogeneous reactions increase with particle acidity. The role of aqueous solution chemistry in organic formation and transformation was also discussed, with an emphasis on the large gap between the complexities of aqueous chemistry and the formation of HUmic LIke Substances (HULIS). The session provoked a lively discussion. A key question was that of the radical conditions in the chamber experiments and how well these represent ambient conditions. Whilst the current data are important for illustrating potential processes and elucidating mechanisms, they may not necessarily be used as direct analogues of atmospheric processes due to the very different oxidant and precursor environments. Recent work in a chamber under near atmospheric conditions shows that the isoprene system appears to have a very low aerosol yield for example. Further studies of this type are required to further test and develop our mechanistic understanding of formation of HULIS in the atmosphere.

There was a detailed discussion of instrumentation for identification of the HULIS and it was recommended that standardised terminology be implemented, since many of the determined parameters are derived and depend on the method of measurements, and that standardised protocols and procedures should be implemented. This is important since as the field develops, results will need to be compared between groups and between different experiments.

The role of aqueous chemistry was also discussed. The question was posed as to whether adding complexity increased understanding of the problem or whether key surrogate systems should be used to identify key processes and rates. A further question related to whether solution phase chemistry was indeed aqueous or whether reaction chemistry took place on the organic phase in the particle.

The talks and discussion highlighted both the importance of humic-like material in atmospheric aerosol, and illustrated its importance for interpreting their cloud activation and radiative properties. The talks also demonstrated what is currently known of its formation pathways and chemical composition but much remains unclear at the time of writing and much work over the coming years will be required to properly ascertain the physical chemistry of this aerosol fraction.

Session 5: How can organic compounds affect atmospheric ice nucleation processes and the properties of cirrus clouds?

The introduction for this section was given by **R. Griffin**, who discussed the climatic importance of cirrus clouds, and defined several mechanisms of ice nucleation in the (UTLS). He also gave examples of recent theoretical, laboratory, and field work investigating the role of organic material

in ice nucleation, and proposed a list of important unresolved issues in this field. These issues include:

- an assumption of water equilibrium in the current model of homogenous freezing;
- a lack of understanding of the chemical nature of organic material in the UTLS;
- a deficiency of knowledge as to which species act as efficient ice nuclei (IN) and what the phase/morphology of particles containing these species is in the UTLS;
- a dearth of thermodynamic data on inorganic-organic-water solutions at temperatures relevant to the UTLS;
- an unknown effect of organic surface coatings on freezing processes;
- the inability to investigate IN without disturbing the ice crystal; and
- the difficulty associated with modelling all of these processes while attempting to understand their relevance for climate.

The homogeneous ice nucleation rate can be parameterised using water activity, independently of whether inorganic or organic solutes are present in aqueous droplets. This theory assumes water is in equilibrium between the condensed and vapour phases. The freezing rate, however, also depends on the volume of the droplets. Because organic particles typically are less hygroscopic than inorganic ones at the high relative humidities (RH) relevant for freezing, organic particles stay smaller. Therefore, in the case of an external mixture of organic and inorganic aerosols, the organic aerosol particles exhibit nucleation rates that are slightly smaller than those of inorganic aerosol particles. Hence, organics are expected to be depleted (with respect to interstitial aerosol) in cirrus ice particles, in agreement with observations. However, **T. Koop** showed model results that indicate this has a negligible effect on the properties of the cirrus clouds.

Moving to the topic of organic surfactants, T. Koop speculated that these substances significantly reduce the accommodation coefficient of water onto organic-containing aerosols. Such a change may strongly influence the homogeneous freezing properties of such aerosols and, more importantly, the properties of the resulting cirrus clouds, *e.g.*, their ice particle number densities. Self-assembled monolayers of long-chain alcohols can also act as very efficient IN in the laboratory. Finally, recent laboratory experiments suggest that organic acids are inefficient ice nuclei (IN) in this deposition mode. Rather, crystalline particles of these acids undergo deliquescence, even at conditions highly supersaturated with respect to ice. It appears that immersion freezing is the most likely pathway for heterogeneous ice formation in the UTLS. Thermodynamic data suggest that mixtures of organics have much smaller deliquescence RH than pure organics, suggesting that efflorescence of such particles in the atmosphere is unlikely. In laboratory measurements, however, organics might crystallize heterogeneously on cirrus ice particles in a first cloud event, opening the possibility that they act as IN during a second cooling cycle.

Because immersion freezing appears to be the most likely pathway for heterogeneous ice nucleation in the UTLS, its relative importance of heterogeneous ice nucleation depends on the presence of solid organic inclusions. **T. Peter** presented several studies that have indicated the presence of organic acids (oxalic, succinic, umaric, phthalic, *etc.*) in particles above their aqueous solubilities, indicating the likelihood of forming a separate phase. In the UTLS, however, it has been shown that organics are likely to be internally well mixed. In addition, as mixtures become more and more complex, the deliquescence RH decreases, meaning that the probability of a particle being crystalline decreases considerably. An exception to these general rules appears to be oxalic acid, which can form crystalline oxalic acid dehydrate (OAD, $(COOH)_2$ $2H_2O$). Using Particle Analysis by Laser Mass Spectrometry (PALMS), particles in the UTLS have been shown to be enriched, in some cases, in oxalic acid. The ability of oxalic acid to act as an IN was studied in the laboratory, and it was found that a shift in freezing temperatures to warmer values was observed for mixtures containing crystalline OAD. This was not the case for other organic acids. In addition, it was found that OAD grows at the expense of liquid particles (a Bergeron-Findeisen effect), further supporting the hypothesis that OAD can be found in crystalline particles and act as immersion IN in the UTLS. All of the data concerning OAD acting as an IN in the UTLS was incorporated into the global ECHAM4 climate model, with the result being a net overall global cooling effect of -0.3 W m^{-2}. This is caused by an increase in high-altitude cirrus that have an approximately 20% decrease in optical thickness.

P. Rairoux showed how ice particles in different types of cloud exhibit various shapes and sizes, as observed by the Cloud Particle Imager (CPI). The ice particle shapes and sizes depend on altitude, with size and complexity of shape generally decreasing with increasing altitude. It is clear that shape and size affect the optical properties (and therefore the associated climate effect) of ice particles due to the different modes of scattered light, but it is doubtful that the presence of any organic material would have a similar effect. An example of geometric optic phenomena is halo formation resulting from small frozen ice particles, even though it is significant ly easier for a large particle to freeze. Remote sensing techniques, including those that are satellite based, for detection of both visible and sub-visible clouds are a means by which future measurement of cloud and ice properties may be improved. In addition, it was speculated that optical techniques currently used for ice particle and cloud characterization/detection could be adapted to focus on characterization of IN inclusions within ice particles. This would allow IN characterization without disturbing the ice particle. Ice particle size and shape are relevant to climate issues because they affect the planetary radiative balance/forcing, and quantities such as the single scattering albedo.

Discussion following these presentations focused on organic surfactants and whether monolayers derived from such species persist long enough in the atmosphere to play a significant role in ice nucleation, the climate relevance (an estimated cooling effect with a magnitude of 0.3 W m^{-2} versus the global cloud cooling effect of 20 W m^{-2}) and terminology (climate "forcing" versus climate "effect") of oxalic acid acting as IN in cirrus clouds, and the potential use of optical techniques for characterization of IN in cirrus ice particles without disturbing the ice particles themselves.

Session 6: How well is the reactive uptake of (organic) gases by an organic and/or ice surface quantified/understood?

This session was introduced by

M. Rossi and started off with a presentation by **S. Borrmann** on recent field studies on the uptake of organics in cloud droplets, interstitial aerosols, cloud residual particles, and ice clouds. One of the main instruments under development for aerosol field studies is an airborne version of the aerosol mass spectrometer (AMS), which is a flightworthy variant of existing AMS instruments in use for ground measurements of the chemical composition of boundary layer aerosols. The quantification of the sulfate and ammonium content of aerosol particles is usually straightforward, but the determination of the nitrate content is more uncertain. The distinction between internal and external mixtures in atmospheric aerosols is of high relevance because the mixing state determines the optical properties of the aerosol that will have direct ramifications on the climate activity of such aerosol particles. Owing to the limitation of the thermal or photothermal (laser) evaporation of the aerosol particles, the detection of transition metals, mineral dust and elemental (black) carbon in aerosols has not been successful so far. It is estimated that a different measurement strategy will have to be chosen in order to enable the detection of these fractions. Considerable effort has been spent on the airborne virtual counterflow impactor inlet, which has not yet become part of routine measurement. Examples of measurements of aerosols in the presence of warm clouds in Puerto Rico and Sweden, among others, were presented and discussed.

The aging of organic particles by ozonolysis is the transformation of organic aerosols from hydrophobic to hydrophilic. The process takes place on average within 0.7 to 1.2 days and may be attributed to the heterogeneous reaction of O_3, OH and NO_3. Oleic acid, a C_{18} mono-unsaturated carboxylic acid, has served as the model system of choice in studies of ozone-olefin interaction. A key question is the relationship between chemical composition of aged organic aerosol and its cloud condensation nucleation (CCN) properties. **S. Martin** presented measurements of aerosol chemical composition and CCN activity that were simultaneously probed using an AMS and CCN counter, respectively, allowing the identification of specific CCN active compounds in the aerosol as it undergoes ozonolysis. At 350 ppm O_3 and at doses up to 0.1 atm·s of O_3 no CCN activation was observed for 200 nm diameter oleic acid particles at 0.6% supersaturation. The oleic acid had completely reacted away. However, a sharp increase in CCN activity of oleic acid has been measured for O_3 doses greater than 0.1 atm·s, while AMS analysis showed that azelaic acid, a C_9 dicarboxylic acid, increased at the expense of the organic polymer fraction. It thus appears that monomeric material, namely azelaic acid, supports CCN activity in the case of oleic acid aging in the presence of O_3 whereas polymeric material seems to be responsible for the hydrophobic state of aged aerosol but does not activate cloud condensation. In aging studies of methyl oleate in the presence of chemically inert and reactive fatty acids occurring in atmospheric aerosol particles such as dioctyladipate and myristic acid, respectively, transformation of carboxylic acids and other protic compounds to high molecular weight polymeric peroxides has been observed. The reaction mechanism presumably involves reactions of the stabilized Criegee intermediate with the monomeric carboxylic acids.

Urban surfaces are coated with films that both contain organic and inorganic compounds. A typical urban window "grime" contains 94% inorganic ions, 7% of which are nitrates, and 5-10% organics such as dicarboxylic acids, carbohydrates, aromatic hydrocarbons (0.02% of which are polycyclic aromatic hydrocarbons (PAH's)), carbonyls, *etc*. These, as well as organic films on aqueous surfaces, could well be the seat of heterogeneous chemical reactions as atmospheric trace gases are efficiently taken up on organic films in a non-reactive manner. **J. Donaldson** presented studies of anthracene interacting with 1-octanol and oleic acid films. As far as the interaction of ozone with PAH's is concerned a variety of compounds and substrates have been studied such that a coherent picture begins to emerge. The dependence of the reaction rate on the gas phase ozone concentration is always well described using a classical Langmuir-Hinshelwood approach, providing strong evidence that the reaction takes place at the surface, rather than within the bulk of the liquid phase substrates, or the solids. The parameters extracted from the kinetic analysis suggest that the nature of the substrate plays a role in the kinetics, but for constant $[O_3(g)]$ the identity of the substrate affects the rates within about an order of magnitude only. Some aspects of heterogeneous nitrogen oxide photoreactions were discussed in light of observations that NO_2 and HONO evolved from HNO_3 adsorbed on an ice surface upon irradiation with near-UV light. The two products display different wavelength and temperature dependences, suggestive of different sources. Gas phase HNO_3 will adsorb onto acridine at such interfaces. Nitrate has been measured in high concentrations, typically 10% by mass, in urban window grime. These observations, in conjunction with those made on ice surfaces, suggest that HNO_3 could adsorb to organic grime surfaces and undergo photochemical transformation to HONO and NO_2, thereby recycling NO_y to NO_x.

While a full molecular speciation of the organic content of atmospheric particles is almost impossible, it still appears that the major part of the organic fraction of a typical atmospheric aerosol consists of neutral polyoxygenated species or polyols such as catechol/levoglucosan, as well as of mono and dicarboxylic acids. These compounds may interact with different trace gases as shown by **T. Cox** who presented new information about the hydrolysis of N_2O_5 on model aerosols consisting of ammonium sulfate ($(NH_4)_2SO_4$), ammonium bisulfate (NH_4HSO_4), malonic (C_3-dicarboxylic acid), succinic (C_4-) and glutaric (C_5-), as well as mixtures of the foregoing with humic acid. The rate of uptake of N_2O_5 on the above-referenced inorganic aerosols is slightly dependent on $[N_2O_5]$ and is self-inhibited by the presence of nitrate originating from the hydrolysis of N_2O_5, the so-called nitrate effect. The uptake coefficient γ (a parameter describing the rate of these heterogeneous chemical reactions) increases with relative humidity from 5 to 80% with the γ value for malonic acid being distinctly larger than for succinic and glutaric acid. The water content of humic acid aerosol has been measured using FTIR absorption at a given relative humidity, and the uptake coefficient γ is lower by a factor of 20 compared to $(NH_4)_2SO_4$ aerosol at comparable relative humidity. A 6% admixture of humic acid to ammonium sulfate aerosol lowers γ by a factor of two. This significant decrease of γ may be attributed to a phase transition of humic acid and formation of an organic thin film on top of the inorganic core. Experimental evidence points to a thickness of the organic thin film of approximately 15 nm. In conclusion, the surfactant properties of

humic acid are the cause of the significant decrease of the uptake coefficient on such coated atmospheric inorganic aerosols.

Describing heterogeneous and/or multi-phase reactions on such complex susbtrates is a difficult task. **U. Pöschl** presented a kinetic model framework for aerosol and cloud surface chemistry, and gas particle interactions, exemplified by is the oxidation of benzo[a]pyrene (BaP) adsorbed on black carbon, which proceeds on the time scale of tens of minutes or faster compared to the gas phase where lifetimes of PAH's in the presence of ozone are immeasurably long. The molecular structure of the compounds forming the aerosol particle critically determines the theoretical framework of the uptake of atmospheric trace gases using a unified kinetic model based on the additivity of kinetic resistances, depending on the specific layer structure and the elementary processes at hand.

Finally, **M. Pilling** concluded this meeting by summarising the discussions that were held during this event.

Report on the 3rd International Limb Workshop
April 25-28 2006, Montreal, Canada

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This workshop, with 42 people in attendance, was sponsored by SPARC and the CSA and continued the series initiated by Professor **John Burrows** in 2003. The first workshop was held in Bremen and the second was organised by MISU, Stockholm University. The previous workshops have brought together students, new researchers and researchers with extensive experience in the field of limb observations, both in measurements and in the application and interpretation of the observations. The 3rd workshop was no exception to this approach. However, the ever-increasing data base of limb observations is modifying available knowledge and providing clear guidance on both the value and limits of limb observations. The workshop that was ably organised and administered by **Adam Bourassa** at the University of Saskatchewan, included three days of relaxed presentations that provided an atmosphere in which the questions could really probe the understanding of possible problems associated with the instruments and the observations. The final day was used to provide a forum for much more general discussions and met its objective.

The primary inputs to the measurement data base, and to many of the presentations in this workshop came from Odin (both OSIRIS and SMR), Envisat (SCIAMACHY and GOMOS) and from the FTS instrument on the Canadian SciSat-1. Future instruments were also a major item of discussion, in particular the NPOESS ozone mapper and profiler suite, the SWIFT instrument on the CHINOOK mission, as well as possible future instruments/missions such as CATSCAN, TELIS and ALTIUS.

While full details and the actual presentations have all been collected on the OSIRIS web site (http://osirus.usask.ca/osiris/limbworkshopweb/limbworkshop.html) it is of value to look at some of the Workshop highlights and important "recommendations." There is no doubt that limb viewing provides both a mapping and altitude profile product, which greatly extends the limited short term spatial coverage afforded by solar occulters, although GOMOS is attempting to remove this limitation. However, a major uncertainty in the height profile determination is an uncertain altitude registration, and it is clear that this

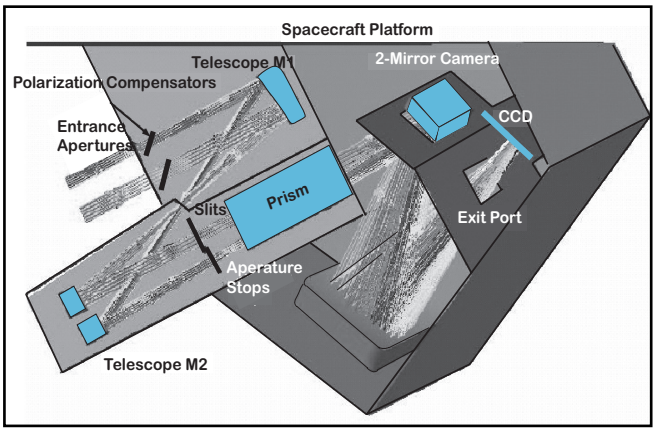

Figure 1: The optical design for the NPOESS ozone profiler instrument. The prism (quartz) spectrometer and 2-d CCD array provide a 290-100 mn, and 2-40 nm bandpass (spectral resolution is matched to ozone absorption features). The polarization compensators minimize sensor sensitivity to polarization.

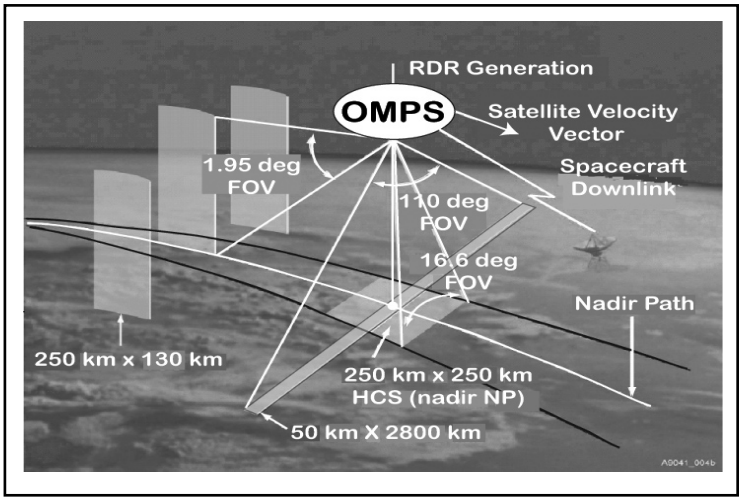

Figure 2: The viewing geometry for the NPOESS ozone profiler instrument (Courtesy Larry Flynn).

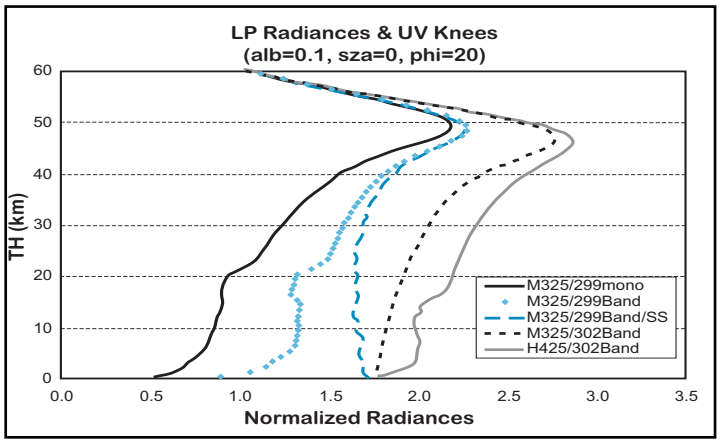

Figure 3: The variation in the knee altitude and the potential uncertainty associated with this method of altitude registration (Courtesy Philippe Xu).

uncertainty is the major contributor to the inaccuracy of the retrieved profiles. The workshop provided a clear discussion of the severe limitations that exist with limb viewing out of the orbit-plane, as it is quite clear that the assumption of horizontal homogeneity in the atmosphere does apply. The possibility of using the limb observations themselves to provide the required altitude registrations was a "hot topic" and it was generally suggested that the required registration knowledge, ~250 m, was simply not possible through this technique. This necessarily implies that independent information is required – star trackers can meet this need but must accommodate any flexure in the spacecraft that will cause a time varying offset for the viewed line of sight.

The other major item of interest to all at the workshop was the problem of scattered light and the out-of-field signal. It is interesting to note that this is not only an optical problem but also extends to microwave and sub-millimeter instruments, in which the side-lobes act in a similar manner. The dynamic range associated with the scattered sunlight limb signal, in the visible, covers six orders of magnitude between 5 km and 100 km tangent height, and can be much larger if the field of view extends to the ground. Thus designing, building and operating instruments that can meet these requirements is not an easy task. The matter of scattered light, or unwanted signal, is also affected by the choice of spectral resolution for the measurement of the scattered sunlight in the limb, so the workshop also considered this point. The NPOESS team is making a considerable effort to develop software solutions to the problem of the unwanted scattered light signal, and to attempt to correct the collected data, but the probability of success is not assured. The OMPS team has also used a prism to defeat the problem of multiple spectral orders that usually present with a grating instrument (**Figure 1**). The impact of polarization on this instrumental approach will not be fully known until this type of instrument is in orbit and making measurements. The basic design of the NPOESS ozone profiler and its viewing geometry are shown in **Figures 1** and **2** respectively. The knees seen in the measured limb radiance are shown in **Figure 3**. The improvements in the quality of the collected data are clearly indicating the need for improved radiative transfer models, and some outstanding advances were presented by young researchers in attendance at the workshop. Perhaps the most important point was the need for true 3-D spherical radiative transfer models. Given the major advances in computing capabilities in the last few years this is not as impractical as once thought (see **Figure 4,** colour plate I). Other interesting topics in the workshop were the presentation of new scientific findings from the limb viewing satellites that are currently in orbit (see **Figures 5 and 6**, colour plates I and II). The effects of solar storms was well identified in the observations of the 2003 Halloween strom by SCIMACHY (see **Figure 7**, colour plate II and III). These are quite significant as a number of United States satellite platforms are not currently operating and so limit the long-term data base that is necessary to study trends.

On the last day of the workshop the participants attempted to identify some essential needs for the future of Limb viewing. The primary one was the packaging of radiative transfer and retrieval codes in order to allow proper testing of the various schemes. One possibility is to use the various retrieval schemes on identical data sets in order to improve our understanding. Indeed, it is hoped that it may be possible to standardize the procedures. The meeting also suggested that priority should be given to the retrieval of scientific products for BrO and aerosol.

The 3rd Workshop benefited from the support of both SPARC and CSA that enabled a number of students to attend. It is hoped that the 4th International Limb Workshop. will occur in the United States in late 2007.

Stratéole-Vorcore : A study of the Antarctic polar vortex using ultra-long duration balloons

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Introduction

The Stratéole project was initially proposed by the LMD (Laboratoire de Météorologie Dynamique) in order to study the dynamics of the Antarctic polar vortex. The first preparatory studies started in 1992, and in 1998 it was decided to separate this project into two independent phases: Vorcore would concentrate on the dynamics of the vortex core, followed by Voredge to study the vortex edge.

The Stratéole-Vorcore campaign finally took place between September-October 2005 from the American base at McMurdo (78°S, 166°E). This base was chosen because it is the only one in Antarctica offering the necessary logistic support and, more importantly, wind conditions at ground level permit stratospheric balloon launches.

The campaign was a joint effort of CNES (Centre National d'Etudes Spatiales: the French Space Agency) and CNRS (Centre National de la Recherche Scientifique). It also benefited from very significant support from the US National Science Foundation as well as from IPEV (Institut Paul Emile Victor: the French polar Institute).

Objectives of the Vorcore experiment

The causes of the formation of the ozone hole are now well known. Air masses inside the polar vortex are isolated from those of mid-latitudes by the winter stratospheric jet. They eventually reach temperatures sufficiently low to permit the formation of Polar Stratospheric Clouds (PSC). It is on the surface of PSCs that chlorine and bromine species are chemically activated and can thus participate in catalytic and photochemical reactions that lead to the destruction of ozone. On the other hand, the permeability of the polar vortex and its capacity to export to lower latitudes air masses that are either poor in ozone and/or chemically activated to destroy ozone are still not well understood.

Thus, the main objective of the Stratéole-Vorcore campaign was to document the austral polar vortex, a region where *in situ* measurements are sparse, in order to study its dynamics from the end of winter, when the ozone hole forms, and up until its final breakdown in spring. In particular, the project was designed

(i) to characterise planetary waves and their role in dispersion and mixing in the vortex,
(ii) to describe shorter-scale gravity waves above Antarctica, and
(iii) to investigate the dynamical isolation of the vortex.

The observing system

The observation system used to reach these objectives was a flotilla of long-duration, closed-envelope, super-pressure balloons (SPB). The pressure of helium inside a SPB is greater than the air pressure of the environment. Thus, they stay on constant density (isopycnal) surfaces. Although their trajectories are not really isentropic, isentropic and isopycnal surfaces are relatively close, and therefore the balloons perform quasi-Lagrangian flights, serving as a good tracer of air parcel motions. If its envelope does not leak, a SPB is able to drift for several months in the lower stratosphere, carrying a light payload. Balloons with either 8.5 or 10-meter diameter were specifically developed by CNES for this campaign. As the envelope is fragile when not fully inflated, a new launching technique was also developed. The balloon is laid on a mobile table and protected by a plastic sheet during ground operations. This sheet is progressively removed in order to keep a well-formed bubble during the inflation.

The scientific payload was designed by LMD. Its weight was 15 kg, including batteries. During the day the gondola was warmed using solar cells while at night the thermal insulation enabled an internal temperature high enough to sustain the electronics.

Every 15 minutes the gondola measured pressure, temperature and GPS-derived position, from which winds are deduced. Data were transmitted to the ground using the ARGOS satellite system.

This working procedure was successfully tested during several preliminary campaigns in Ecuador, Sweden and Brazil. The test flights themselves provided important scientific results. For example, they provided the first quasi-Lagrangian observations of Yanai waves at the Equator and an estimate of gravity wave momentum fluxes in this region (Vial *et al*., 2001; Hertzog and Vial, 2001), and they confirmed the existence of an excess of energy at inertial periods at high latitudes (Hertzog *et al*. 2002). These preliminary observations proved to be very useful in testing the validity of Meteorological Weather Forecast Models (Hertzog *et al*., 2004; Knudsen *et al*., 2006).

The campaign

The operational team, composed of six members from CNES and three members from LMD, arrived in McMurdo on August 20th with the first "winfly" flight from

Christchurch. Four Winflys were scheduled in order to prepare the station for the summer season starting in October. Only thirty scientists arrived on these flights, including Terry Deshler's group (University of Wyoming) who collaborated with the Vorcore team.

On August 20, under severe weather conditions, an American team began to prepare the temporary launch pad on the sea-ice in front of the station. The work included the construction of two "Jamesway" buildings, each about 100 feet long, necessary for preparing the balloon before launch and for setting up the power generators. Most of the hardware (95%) was shipped earlier, arriving at McMurdo in the previous February.

Flight operations began on September 5. One of the Jamesways was partly destroyed by a storm that day, but a NSF team worked very hard to reconstruct it and the launch window was not lost. A total of 27 balloons were released in two months, with the temperature on the ground sometimes reaching as low as –38ºC. The last balloon was automatically destroyed in flight on February 1. The mean lifetime of the gondolas was 63 days, with the longest flight duration being 109 days. More than 150,000 observations were made, corresponding to a total of 1575 days. Except for two balloons that experienced problems during launch and three with leaky envelopes, most balloons had to be automatically destroyed, either because they were drifting northward of 40°S, a limit fixed for security reasons, or because the gondolas were running out of energy. **Figure 1** shows an example trajectory for one of the balloons (flight 3) and the corresponding temperature data obtained from that flight are shown in **Figure 2**.

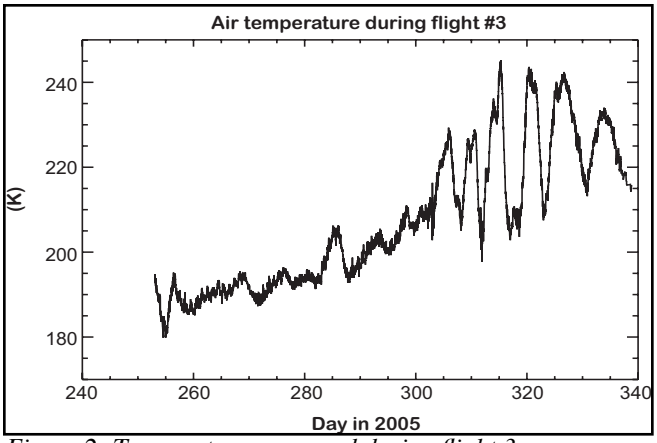

Figure 2: Temperature measured during flight 3.

The geographical sampling shows a tendency for a better documentation of the 60°S-80°S latitudinal band and of the 60°W-120°E longitudinal sector (as the vortex had a tendency to be centered off the Pole). However, the sampling of the vortex is very good everywhere. Thus the Vorcore experiment, the biggest operation made in McMurdo in wintering conditions, can be considered a great technical, operational and scientific success.

After the end of the campaign the Vorcore data base was quality checked and errors corrected. This mainly concerned a small warm bias of temperature measured during day-time. The data base was distributed to participating teams in early April.

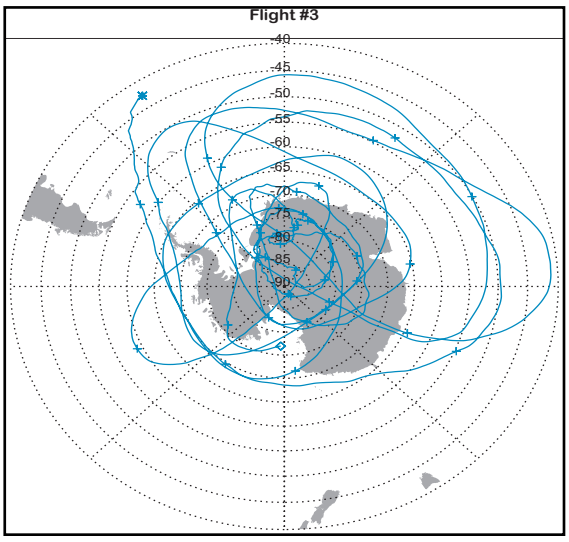

Figure 1: An example of balloon trajectory, flight 3.

Firsts results

Analysis of the data set is now underway.

In the Antarctic Ozone Bulletin No 8/2005, it is noted that the 2005 austral polar vortex was close to the average of the last decade but that the minimum temperatures inside the vortex were, in early September, near the coldest recorded since 1979. Indeed, very low temperatures (down to 180°K) were also observed by the SPBs in early September.

The balloon trajectories within the core of the vortex show a lot of small-scale dynamical structures, which have to be further studied. The trajectories also showed a period of very intense planetary wave activity up to mid December, which was followed by a dynamical regime dominated by gravity waves having periods close to the inertial period. More than 15 balloons were simultaneously afloat from mid October to mid December. This will allow the final breakup of the vortex to be precisely studied.

In mid November, the vortex was pushed away from South Pole. At the end of November one of the balloons was ejected into a potential vorticity filament. In early December the final destruction of the vortex began with distortion by a zonal wavenumber-3 planetary wave, followed by the break up of the vortex and the formation of several smaller vortices, in which several balloons were trapped. These vortices finally disappeared during the establishment of the lower stratospheric summer circulation.

Mountain waves with very large amplitudes (more than 15°K peak-to-peak) were observed on several occasions above the Antarctic Peninsula or above the boundary between the ocean and the continent. Two balloons trapped in such waves were even destroyed by the excess of over-pressure they induced inside the balloon envelope.

Studies underway

This data set will be used to investigate the accuracy of operational weather forecast models in a region where the *in situ* observational network is rather sparse. This is important as it enables any biases in these models to be investigated.

Initially, the focus is on the gravity wave activity. Because the balloons drift following air masses, we can obtain spectra of momentum flux as a function of intrinsic phase speed (see **Figure 3**, colour plate III). This quantity is essential for constraining the parameterization of gravity waves in general circulation models. We have developed a method to determine these fluxes, based on the fact that the SPB drift along isopycnal surfaces. Wave disturbances induce vertical displacement of these surfaces, which can be used to infer the vertical wind fluctuations associated with the waves. To do that, it is necessary to relate the vertical

displacement of the isopycnal surfaces to those of the isentropic surfaces on which air masses move. The vertical displacement of the balloon must be deduced from the pressure fluctuations as the GPS position is not precise enough. However, the measured pressure fluctuation is a combination of two effects: 1) the vertical displacement in the mean pressure gradient and 2) the pressure change due to the wave itself. Hertzog and Vial (2001) assumed that the last term is negligible and were able to study momentum fluxes in the Equatorial lower stratosphere. The new method relaxes this assumption and, using the gravity wave polarization relation, allows a better estimate of the momentum fluxes. Furthermore, it also allows us to deduce the intrinsic phase speed and the wave propagation direction.

Numerical simulations show that the method is robust, although the momentum fluxes are slightly underestimated. The technique will provide a depiction of gravity wave activity as a function of geographical position and time and so it will be possible to study wave sources. Furthermore, regular radiosonde soundings provide a vertical (Eulerian) description of wave activity and will thus be used to complement the horizontal (Lagrangian) description given by Vorcore balloons.

Study of the dispersive properties of the vortex is also being undertaken. This is done by evaluating the separation of a pair of balloons that are initially close to each other as a function of time. The launch strategy during Vorcore was thus designed to launch two balloons in tandem. However, pairs of balloons also formed randomly during the experiment. A preliminary study, made when the experiment was still underway, showed that the mean distance of a pair of balloons after 6 days was 1500 km (2000 km) when they were initially at a distance smaller than 100 km (300 km) respectively and that this distance grew linearly with time (M. Gutiérrez and P. Aceituno private communication). The number of pairs of balloons seems to be large enough to permit a good statistical estimate of this dispersion law. However, these preliminary results have to be confirmed and further analysed. Due to calendar uncertainties, it was impossible to schedule a "Match" experiment (similar to QUOBI (Quantitative Investigations) which took place in 2003 in Antarctica) during Vorcore. The "Match" method allows an estimation of the ozone destruction rate by sampling a given air mass with ozone soundings made at different stations working on alert using trajectory forecasts to decide when and where the soundings should be made. However, several groups that perform ozone soundings in Antarctica (University of Wyoming, Alfred Wegener Institute, Australian Antarctic Division, Finnish Meteorological Institute, Instituto Nacional de Técnica Aeroespacial and Argentinean Meteorological Service) had agreed to make additional soundings on alert when Vorcore balloons were forecast to fly over their stations. With more than 180 overpasses at distances smaller than 100 km over the relevant stations (also including Admussen-Scott, Syowa and Dumont d'Urville), this small "Match-like" campaign is expected to produce results on ozone depletion over Antarctica for 2005. It should also give some feedback to further analysis of the QUOBI campaign.

Balloon 1 just before the launch

The analysis of these soundings and of the SPB trajectories should also provide further insights on ozone laminae entering the Antarctic vortex (Moustaoui *et al.,* 2003), another mechanism inducing a mixing of air masses from different origins. Together with the Vorcore observations, this should permit further studies of the definition of vortex edge as being a zone where the mixing is very low (Lee *et al.,* 2001). With all these studies, we can expect to obtain a better estimate of the vortex permeability.

Last but not least, modelling studies will be important components of future activities. Several groups have agreed to test the impact of the assimilation of Vorcore data on their numerical weather forecasts. Furthermore, the University of Manchester (M. Chipperfield and W. Feng) made runs of the Chemistry-Transport Model SLIMCAT during Vorcore, data of which will be very valuable for interpreting the model outputs.

Further results, including an animation of each balloon's trajectory, can be found on the Vorcore website: http://www.lmd.polytechnique.fr/VORCORE/McMurdo.htm.

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A new web-based resource for studying major, mid-winter stratospheric sudden warmings

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Introduction

The last few years have seen a renewed interest studying and classifying major, midwinter, stratospheric sudden warmings (Limpasuvan *et al.* (2004); Manney *et al.* (2005); Charlton and Polvani (2006)). This has been motivated, in no small part, by the recognition that the dynamical connection between the stratosphere and troposphere is particularly strong during periods of large stratospheric disturbance (*e.g.* Baldwin and Dunkerton (2001)). In fact, coupling between the stratosphere and troposphere is one of the three SPARC scientific themes (see O'Neill and Ravishankara (2004)) and, more and more, the stratosphere and troposphere are being understood as two components of one tightly coupled system. A recent review of relevant questions may be found in a previous newsletter article (Haynes, 2005).

An illuminating analogy, we suggest, may be drawn between stratospheric sudden warmings (SSWs) within the stratosphere-troposphere system, on the one hand, and the El-Niño Southern Oscillation phenomenon within the atmosphere-ocean system, on the other. For each system these phenomena offer the most prominent instance of the coupling between the two components. As such, understanding these key phenomena would shed much light on the specific nature of the coupling, and would likely be important in successfully modelling the fully coupled systems.

SPARC has a long tradition of studying SSWs and, through the SPARC Newsletter, of informing the community about their occurrence, dynamics and consequences. In particular the work of **Karin Labitzke** and her collaborators in Berlin has provided extensive records of SSW activity between 1952 and the present time (*e.g.* Labitzke and Naujokat (2000)). In this article we introduce a new web-based resource which, we hope, will be of help to the community of scientists interested understanding the detailed dynamics of SSW events and, more generally, the coupling between the stratosphere and the troposphere.

The new website offers many diagnostic quantities for each SSW event since 1958, as well as animations of the potential vorticity field at various heights in the stratosphere, and even three-dimensional renderings of the evolution of the vortex shape during the events.

quantity	levels	type	time range	position
zonal mean zonal wind	10 hPa	static	NDJFM	at 60 N
polar cap temperature	10 hPa	static	NDJFM	average 50-90N
meridional heat flux	100 hPa	static	NDJFM	average 45-75N
geopotential height	10 and 100 hPa	animation	daily, +/-30 days	map
potential vorticity	450 K, 840 K and 1200 K	animation	6 hourly, days -9 to +5	map
3d PV isosurfaces	450 K to 1600K	animation	6 hourly, days -9 to +5	map

Table 1: Diagnostics available, for each SSW event, on the new website.

We have designed, tested and published the new website using both the NCEP (Kalnay *et al.*, 1996) and ERA-40 (Kallberg *et al.*, 2004) reanalysis data sets. The website can be found from links on the SPARC Data Center webpage, or directly at the following URL: **http://www.appmath.columbia.edu/ssws**. In the sections below, we first explain how the SSW events on the new website are selected, we then detail which specific diagnostics are available at the website, and we finally illustrate the capabilities of the website by considering a specific SSW event in some detail.

Methods

One of the novelties of the new website is that the SSW events described there were extracted from the reanalyses with an algorithm that automatically scans the data sets and detects the events. An important advantage of this procedure, beyond eliminating the inevitable biases due to human intervention, is that it can be also applied, with no modification, to model output. This allows for easy evaluation of model output versus the reanalyses (Charlton *et al.*, 2006). The algorithm used to detect SSWs is only briefly sketched here, and the full details can be found in Charlton and Polvani (2006). As a general principle, we have designed the algorithm so as to keep as close as possible to the standard WMO definition of SSWs, but readers should note that there are some differences between our definition and that of Labitzke (1977). Three condition are required for an SSW to be defined:

- the zonal mean zonal winds at 60ºN and 10 hPa must become easterly (during the months NDJFM), and
- no other SSW must have occurred for the preceding 20 days, and
- following the SSW, the zonal mean zonal winds at 60ºN and 10 hPa

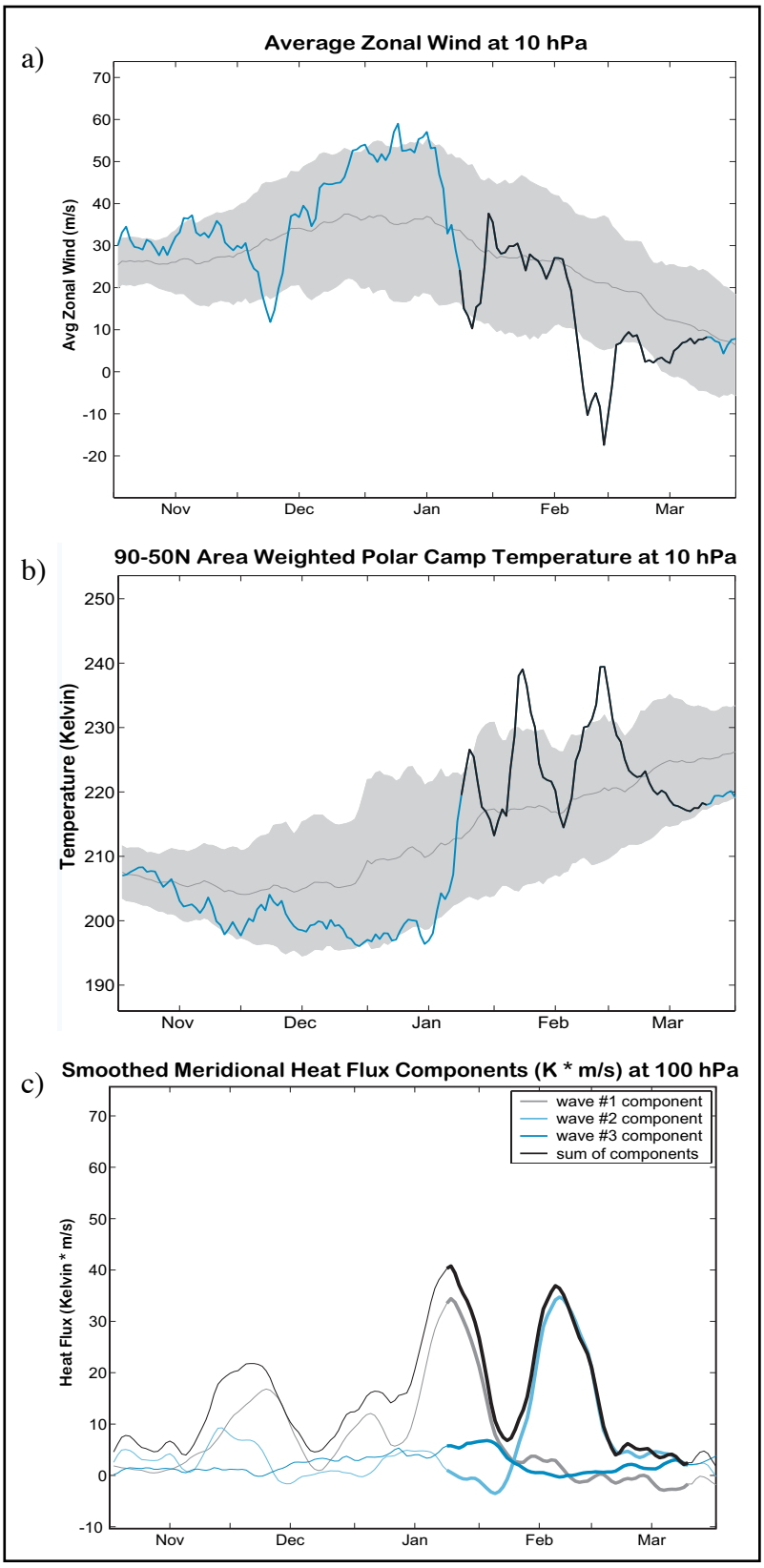

Figure 1: (a) Zonal mean zonal wind at 60ºN and 10 hPa for winter season 1978-79 (blue line). The black line shows period ± 30 days around the central date of the SSW event. The grey line shows climatology, 1958-2001, and grey shading shows climatology ± one standard deviation. (b) as (a) for area weighted polar cap temperature 90-50ºN at 10 hPa (c) Smoothed meridional heatflux (45-75ºN) at 100 hPa for winter season 1978-79. The black line shows total heatflux, the grey line shows wavenumber 1 component, the light blue line shows wavenumber 2 component, and blue line shows wavenumber three component.

must become Westerly for at least 10 consecutive days before April 30th of that winter season.

The first criterion is self-explanatory, the second one is meant to avoid double counting of events, and the third one is designed to exclude final warmings. With this definition we have analysed the NCEP and ERA-40 reanalysis data sets, for their common period, winter season 1958/59 to winter season 2001/02. Each SSW event that was found has been labelled with a date corresponding to the first day of observed easterly winds at 60ºN and 10 hPa; we refer to this as the central date. We have found 27 SSWs in the NCEP data set and 29 in the ERA-40 data set, most of which are common to both data sets. For more details about the climatology of these events, see Charlton and Polvani (2006).

Diagnostics

For each SSW, we have calculated a set of common diagnostics and have plotted them on similar graphs for easy comparison. We next describe some of the diagnostics available on the site, as summarised in **Table 1**. They fall into two sets: the first comprises several familiar Eulerian mean quantities that are commonly used to analyse SSW events. The second set of diagnostics focuses on potential vorticity (PV), and offers both contour maps of PV on isentropic surfaces and 3-dimensional renderings of the whole vortex structure. This second set of PV diagnostics is available for the ERA-40 data set only. Also, for each event, we have produced animations of both the geopotential and PV fields; we trust this will help provide further insights into the evolution of the polar vortex during SSW events.

a. Zonal Mean Zonal Wind

The zonal mean zonal wind at 10 hPa and 60ºN is plotted for the winter season in which each SSW occurs, together with the mean winter climatology one standard deviation (derived using all the winter seasons from 1958/59 to 2001/02). This diagnostic is useful to determine the amplitude of the wind deceleration following each event.

b. Polar Cap Temperature

The area-weighted average (between 90-50ºN) of temperature on the 10 hPa pressure surface is plotted for the winter season in which the SSW occurs. As in the zonal wind plot, the climatology and variability are also shown.

c. Meridional Heatflux

The meridional heatflux, a proxy for the vertical component of the Eliassen-Palm flux, averaged between 45-75ºN, is plotted for the winter season in which the SSW occurs. Again, both the climatology and the variability are graphically illustrated. In addition to the raw meridional heatflux, we have produced a smoothed ver-

sion (with a 15-day running mean filter applied), the individual components for each longitudinal wave numbers and, following Polvani and Waugh (2004), the integral for 40 days prior to each day.

d. Geopotential height maps and animations

Polar stereographic plots of geopotential height are available, with 12 hour resolution, from 30 days before to 30 days after each SSW event. The plots are available on both the 10 hPa and the 100 hPa levels. An animation tool was developed to allow the user to play the animation backwards and forwards, change the frame-rate of the animation, step through each event frame-by-frame, and jump to a specified frame.

e. Isentropic PV maps and Animations

Polar stereographic projections of potential vorticity on the 450, 840 and 1200 K surfaces are plotted from 9 days before to 5 days after the central date of the SSW, with a time resolution of 6 hours. In these plots, the modified PV (see below for details) is used instead of the Ertel PV: the latter can easily be transformed back to Ertel using a simple multiplication factor. Animations are available at each of the levels.

f. 3D Isosurfaces of PV and Animations

Three dimensional isosurface plots, and the corresponding animations, of the vortex edge are available between 9 days before and 5 days after the central warming date, with six hourly resolution. In order to show a meaningful plot from the 450 K to the 1600 K levels, the PV is rescaled as suggested by Lait (1994) and in the spirit of Polvani and Saravanan (2000). The isosurfaces are constructed using a single value of modified PV, chosen to represent the vortex edge, and selected 30 days before the central warming date; this value is fixed for each event.

An example

In this section we offer an illustration of the diagnostics available on the new website in the context of one particular SSW event, in February 1979. This event has been extensively studied by a number of authors *(e.g.* Manney *et al.* (1993); Mechoso *et al.* (1984); Simmons and Strufing (1983); Palmer (1981); Dunkerton and Delisi (1986)), and thus will be familiar to many readers. Only ERA-40 diagnostics are used in this discussion.

The zonal mean zonal wind profile for the winter season 1978-79 is shown in **Figure 1a.** The zonal mean zonal wind profile is typical of winters in which late vortex splitting SSWs occur; note the early minor disturbance to the vortex in mid-December. A minor warming occurs in January, followed by the major warming in late February. The rapid deceleration of the zonal mean zonal wind is accompanied by rapid rises in the polar cap temperature, which also lies far outside the normal variability for this point in the season, as can be seen in **Figure 1b**.

The SSW is also characterised by a burst of meridional heatflux in the wavenumber 2 component and, as has been noted in several previous studies, that event is preceded by a minor warming which is associated with large meridional heatflux in the wavenumber 1 component (see **Figure 1c**).

A synoptic view of the SSW is shown in **Figure 2** (see colour plate IV). The left column shows the geopotential height at the 100 hPa level. The minor warming in January is clearly seen as a displacement of the polar vortex off the pole, whereas the major warming in February is a clear vortex splitting event, with approximately a third of the vortex material drawn into the secondary vortex over North America which eventually spins down leaving a much smaller polar vortex in March than normal.

The PV on the 840 K potential temperature surface (**Figure 2**, middle column) shows the same splitting of the polar vortex into two large reservoirs of high PV air. Note that, as the vortex splits, low PV air from tropical latitudes is drawn around the vortex in a thin streamer and then over the pole as the vortex splits.

A three-dimensional perspective of this event, shown in the right column of **Figure 2**, allows one to view the time development of the split in more detail. Until just before the split in the middle stratosphere there is little evidence of dynamical activity in the structure of the vortex other than its tilt southward with height and slightly small size. When the split does occur in the middle stratosphere, it does so relatively rapidly until two deep columnar, vertically aligned vortices are produced.

Summary

In this brief article, we have described and illustrated a new web-based resource for the study of stratospheric sudden warmings. This new tool will allow detailed comparison between various SSWs, and will lead to greater understanding of their similarities and differences, and assist the design of experiments to examine their dynamics. We hope that it also will enable a closer scrutiny of stratospheric variability, and its role in influencing the tropospheric state.

As mentioned above, we welcome feedback and suggestions for improvement. Please visit the site and send us feedback through its comment button, or directly by email to any of the authors of this short note. We look forward to your thoughts so that we might make this resource more useful in the future.

Acknowledgements

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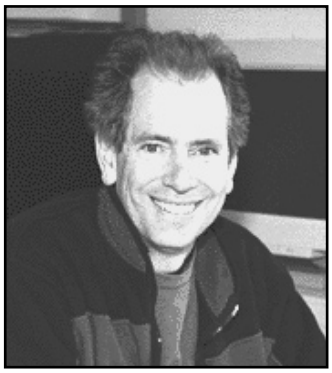
In memory of Byron Boville 1954-2006



We are sorry to announce the death of Byron Boville of cancer at the age of 52.

Byron was a senior scientist within the Climate Modeling Section of the Climate and Global Dynamics Division at the National Center for Atmospheric Research (NCAR) in Boulder Colorado.

He was born in Montreal, Quebec to Byron Walter (Barney) Boville and Grace Mary Jolly. Barney was a professor of meteorology at McGill University.

Byron received a B.Sc. (First Class Honours) in Meteorology from McGill University in 1975. He then moved to Seattle to attend graduate school at the University of Washington under Jim Holton. His PhD thesis topic was on baroclinic wave vacillations, and he was one of the first to demonstrate that changes in the planetary scale circulations of the winter stratosphere can produce significant changes in the troposphere, affecting both stationary and transient eddies. That work has served as a strong influence to many in studying stratosphere/troposphere interactions. The interaction between the middle and lower atmosphere remains an active area in weather and climate studies, and was a topic that Byron remained passionate about throughout his life.

In 1979 Byron received a postgraduate fellowship in the NCAR Advanced Study Program. At that time he started to broaden his interests to explore many other aspects of the atmospheric general circulation. When his ASP fellowship was complete Byron was offered a position as a research scientist at NCAR. He remained a research scientist (at increasing levels of seniority and responsibility) at NCAR for the remainder of his life.

Although originally trained in large scale fluid dynamics, his interests and knowledge extended far beyond that area. He worked knowledgeably and competently in climate detection, atmospheric tracer transport, boundary layer turbulence, and numerical methods, and was equally comfortable in the technical fields of software engineering, algorithm design, and parallel methods for supercomputers. His range of expertise made him a general circulation modeller "par excellence", and the vast number of disciplines where he worked comfortably helps to explain his key role in the development of: 1) the "Whole Atmosphere Community Climate Model (WACCM)," a comprehensive model that incorporates physical and chemical processes required to investigate coupling between atmospheric regions from the surface to 150 km; and 2) the Community Climate Systems Model (CCSM), a comprehensive model of the tropospheric climate system that encompasses many of the physical, chemical, and biogeochemical components important for studies of climate and climate change. **Byron received the CCSM Distinguished Achievement Award of 2006 for his contribution to the development of general circulation models, and for the original and innovative thinking with which he challenged himself and his colleagues during his distinguished career in NCAR.**

In addition to publishing regularly in the peer reviewed literature, his expertise in these many areas, and his energy, and volubility made him a natural for serving on numerous national and international advisory panels, and in authoring chapters, and reports throughout his career.

Byron was also an avid sportsman, and participated in athletic activities with the same enthusiasm as he did with science projects. He was a fanatical whitewater kayaker and loved to introduce the sport to friends and family and to be a part of subsequent river trips.

We will all remember Byron for his love for life, his care as a friend, his enthusiasm for new ideas, and the focus and rigor that characterised all his work. We will miss him greatly.
Phil Rasch, Fabrizio Sassi and James J. Hack

Future SPARC and SPARC-related Meetings

2006

16-23 July: **36th COSPAR Scientific Assembly and Associated Events**, Beijing, China (http://www.cospar2006.org)

17-23 September: **Joint IGAC/CACGP/SOLAS/WMO Symposium: Atmospheric Chemistry at the Interfaces - The 9th Scientific Conference of the IGAC Project**, Cape Town, South Africa

26-28 September: **ADM-Aeolus Workshop,** ESTEC (European Space research and Technology Centre), Noordwijk, The Netherlands (http://www.congrex.nl/06c05/)

2-4 October: **SPARC Data Assimilation Workshop**, ESTEC, Noordwijk, The Netherlands (http://www.atmosp.physics.utoronto.ca/SPARC/DA/DAhome.html)

4-6 October: **First SOLARIS meeting**, Boulder, CO, USA http://strat-www.met.fu-berlin.de/~matthes/sparc/solaris.html

9-12 November: **Global Environmental Change: Regional Challenges, An Earth System Science Partnership Open Science Conference**, Beijing, China

1–15 December: **AGU Fall Meeting**, San Francisco, CA, USA

2007

14-18 January: **AMS Annual Meeting, San Antonio, Texas, USA**

* 19th Conference on Climate Variability and Change,
* 11th Conference on Integrated Observing and Assimilation Systems for Atmosphere, Oceans, and Land Surface (IOAS-AOLS),
* 11th Symposium on Meteorological Observation and Instrumentation,
* 9th Conference on Atmospheric Chemistry.

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