Lower Stratosphere
NERC UTLS OZONE
Upper Troposphere

UTLS OZONE Newsletter

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Editor: R. Penkett

UTLS OZONE Mission Statement

The UTLS OZONE Programme aims to advance scientific knowledge about middle latitude ozone and its role in the Earth's climate system, thereby enabling authoritative statements to be made about ozone change. This will be achieved by quantifying the impact of human activity within the context of natural variability.

Welcome to the third edition of the Upper Troposphere Lower Stratosphere (UTLS) OZONE Newsletter. The UTLS OZONE thematic programme has now been running for three years. A timetable of forthcoming events can be found below.

Future activities	2001-2002
UTLS Field Campaign Data Workshop - Cambridge	17th-19th December 2001
Science Meeting - Bristol	8th-11th July 2002

Over the last year, UTLS OZONE scientists have been very active collecting new data on the composition of the UTLS and increasing our understanding about this region of the atmosphere. In July 2000, a Strategy Meeting was held to review progress made so far and to identify gaps in the research programme. A summary from this meeting can be found on the web site (http://utls.nerc.ac.uk). Three areas were identified as important research topics, which were targeted in the fourth UTLS OZONE funding round (deadline in November 2000). These were aviation impact, chemistry-climate interactions and analysis of existing data, particularly that collected as part of UTLS OZONE projects. A list of successful fourth round projects can be found inside the back cover of this newsletter.

In order to stimulate research proposals in the area of aviation impact and collaboration between scientists and users, a two-day workshop on Aviation Impact Studies was held at the Royal Aeronautical Society (RAeS) in October 2000. The workshop was deemed a great success involving over 40 participants from universities, industry, government departments and non-governmental organisations. The RAeS and Virgin Atlantic Airways also offered their support for this event and they are thanked for their contributions. The Workshop resulted in several new proposals being submitted to the fourth funding round. It also spearheaded a new initiative to equip British commercial aircraft with sensors to measure a range of trace gases. This exciting initiative is now moving forward with the help of the UK Foresight Action 'Greener by Design' Aviation Environment group, British airlines and government representatives. The aim is to complement and to build on existing activities such as the EC MOZAIC and CARIBIC programmes.

In this Newsletter, you will find a series of articles describing results from the fourth UTLS OZONE Science Meeting which was held at the University of Leicester over three days in December 2000. Many interesting results were presented at the meeting showing that the Programme is certainly contributing to improving our knowledge about the UTLS region. There were also several presentations from invited speakers which stimulated lively discussions. Abstracts from some of these talks are included here. An open poster session was also held, again showing some of the many interesting results emerging from the UTLS OZONE Programme.

The next UTLS OZONE Science Meeting will move to a summer slot in June 2002. It will be held at the University of Bristol. Prior to this, a workshop will be held to discuss results from field campaigns funded by UTLS OZONE within the framework of the themes and objectives laid out in the UTLS OZONE Science and Implementation Plans. The aim will be to bring together scientists working on the analysis of data in the UTLS region. The workshop will take place from the 17th to 19th December 2001 at Madingley Hall, Cambridge. Further details will be sent out soon. It is likely that other workshops will be held in the future on, for example, chemistry-climate interactions. If you have suggestions for other topics please let us know.

Finally, we would like to thank Helen Rogers, European Ozone Research Coordinating Unit (EORCU) for all her help over the last year, particularly as temporary Programme Manager (October 1999 to April 2000) and with the organisation of the Aviation Workshop.

Dr Kathy Law and Miss Rebecca Penkett

NATURAL ENVIRONMENT RESEARCH COUNCIL

"Jacqueline" gondola, for a long-duration MIR flight, containing the University of Cambridge ozone instrument, Consiglio Nazionale delle Richerche LABS instrument, and Laboratoire de Meteorologie Dynamique RUMBA instrument (beneath main gondola). UTLS OZONE provided travelling expenses for University of Cambridge researchers for this campaign. Courtesy of Miss K. Turnbull, University of Cambridge.

The Egrett aircraft with project scientists, engineers and pilot after the final flight on the 6th June 2000. Courtesy of Dr J. Whiteway, University of Wales, Aberystwyth.

Instrumentation to measure nitrogen oxide species (NO, NO_2, HNO_3 and NO_y) on-board the C-130 aircraft during the Atmospheric Chemistry and Transport of Ozone (ACTO) and European Export of Precursors of Ozone by Longrange Transport (EXPORT) field campaigns in 2000. Courtesy of Dr N. Brough, University of East Anglia.

Articles from the Fourth UTLS OZONE Science Meeting: University of Leicester, December 2000

The determination of wavelength-dependent quantum yields for acetone photolysis

Acetone, $CH_3C(O)CH_3$, is probably one of the most abundant oxygenated hydrocarbons in the atmosphere. Its fate is controlled predominantly either via reaction with the hydroxyl radical, OH:

$$OH + CH_3C(O)CH_3 \rightarrow H_2O + CH_2C(O)CH_3 \qquad R1$$

or by sunlight (λ > 290nm, ozone cut-off) induced dissociation:

$$CH_3C(O)CH_3 + h\nu \rightarrow CH_3CO + CH_3 \qquad R2$$

The impact of acetone in the atmosphere depends on the ratio of R1 to R2; reaction R1 produces only one radical while process R2 produces two. The rate constant for reaction R1 has been accurately determined but the rate of photodissociation, R2, is complicated as the yield of radicals is pressure dependent. The excited states of acetone from which dissociation occurs are collisionally quenched back to the ground-state in competition with bond cleavage:

$$CH_3C(O)CH_3 + h\nu \rightarrow [CH_3C(O)CH_3]^* \rightarrow CH_3CO + CH_3$$

$$\downarrow [N_2 \text{ or } O_2]$$

$$CH_3C(O)CH_3$$

This pressure dependence becomes more significant at longer wavelengths where the solar flux is increasing rapidly, and under conditions relevant to the atmosphere the quantum yield for photodissociation has been characterised only in a few studies using end product analysis. The work funded by UTLS OZONE represents the first attempt to directly monitor the products from acetone photolysis using a novel methodology. A tunable dye laser is used to photolyse acetone between 275-330nm, and a second tunable dye laser is used to probe the photoproducts. CH_3 and CH_3CO are not readily amenable to detection themselves, but in the presence of oxygen (O_2) they react to generate species that can be probed. In particular, the reaction between acetyl and oxygen produces OH:

$$CH_3CO + O_2 \rightarrow OH + \text{other products (probably formaldehyde + CO)}$$

OH can be probed with great sensitivity using laser-induced fluorescence (LIF), and hence acts as a good marker for the extent of acetone photolysis. When using the LIF it is difficult to relate the fluorescence signal to the absolute OH radical concentration. To circumvent this problem the fluorescence signal at the wavelength of interest is compared to the fluorescence at a reference wavelength (248nm), at which the quantum yield for acetone photolysis is one, and is not pressure dependent. Figure 1 shows the OH fluorescence signal at 310.5nm and at the reference wavelength (248nm) for two different pressures in air. The pressure dependence following 310.5nm photolysis is clearly seen. A Stern-Volmer relationship is found to adequately describe the pressure dependence of the OH yield:

$$1 / [OH] = 1 + k_{SV} [Air]$$

where k_{SV} is the Stern-Volmer rate coefficient for quenching, which enables the quantum yield at atmospheric pressure to be obtained by extrapolation. Figure 2 shows the quantum yield (results are preliminary) at atmospheric pressure for the wavelengths studied to date. In the UTLS region the yields will be somewhat higher.

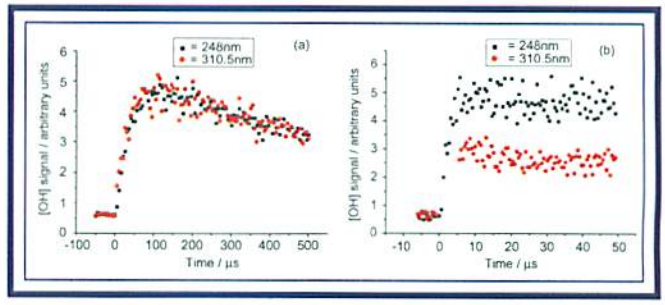

Figure 1

The temporal evolution of OH produced following acetone photolysis in air at (a) 6 Torr and (b) 33 Torr. The OH is generated by the $CH_3CO + O_2$ reaction. Comparison between the yield of OH at the wavelength of interest and 248nm enables the reduction in quantum yield due to collisional quenching of acetone to be determined. The use of the reference wavelength enables the pressure dependence of the yield of OH from the CH_3CO+O_2 reaction to be taken into account.

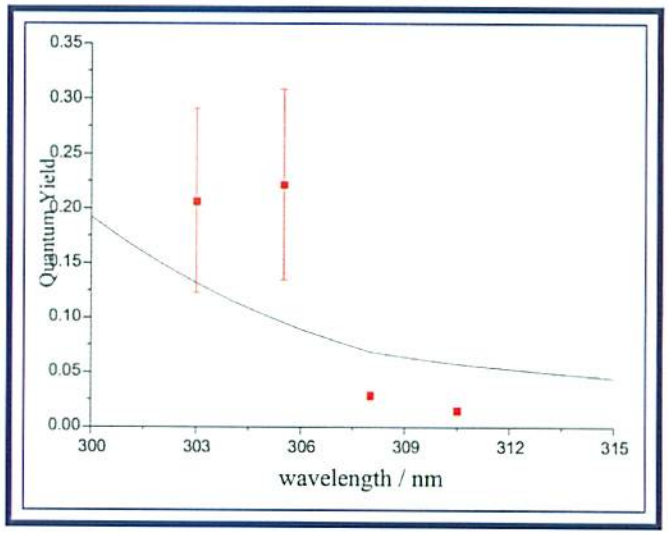

Figure 2

Photodissociation quantum yield for the photolysis of acetone in air at 760 Torr and 298K as a function of wavelength. The bold line represents the results of a previous indirect end product study.

Mark A. Blitz, Dwayne E. Heard and Mike J. Pilling,
School of Chemistry, University of Leeds.

Characteristics of stratospheric air during the drying phase of the tape recorder

The Upper Atmosphere Research Satellite (UARS) was launched on 19th September 1991 to make measurements of a variety of atmospheric constituents. The Microwave Limb Sounder (MLS), an instrument on UARS, is sensitive to water vapour and ozone in the lower stratosphere and made coincident, daily measurements of the two species in the tropical region until April 1993. The Cryogenic Limb Array Etalon Spectrometer (CLAES), another instrument on UARS has a similar spatial and temporal coverage to that of MLS and can be used to indicate the presence of cirrus clouds [Mergenthaler et al., 1999]. We have explored the relationship among these three fields to examine stratosphere-troposphere exchange and the role of convection in dehydrating the lower stratosphere.

Using CLAES data and following the method of Mergenthaler et al. [1999] we searched for cirrus clouds at 68hPa. For each cloud hit, we calculated the corresponding relative humidity value using the nearest MLS water vapour mixing ratio and the European Centre for Medium Range Weather Forecast (ECMWF) temperature at the nearest grid-point and synoptic time to the MLS footprint.

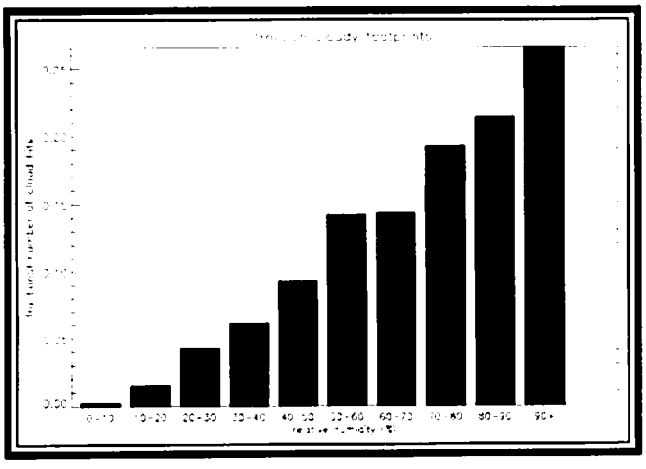

Figure 1
Histogram of the number of CLAES cirrus cloud hits occurring in 10% relative humidity bins at 68hPa during DJF 1992.

The histogram in Figure 1 shows the frequency of cirrus clouds occurring in 10% relative humidity bins at 68hPa during Northern Hemisphere winter in 1992. The frequency of cloud hits increases with relative humidity with most clouds being found in air which is saturated. It can also be shown that more cirrus are found in regions where the temperature is 188-190K, the coldest temperatures found at this level at this time. A similar analysis to examine the distribution of cirrus within the MLS ozone field at 68hPa leads to Figure 2. Ozone mixing ratios have a tendency to be lower in regions where cirrus clouds are present which may indicate that the air in which cirrus clouds are found is tropospheric in origin or that ozone is destroyed on ice particles within the clouds.

In the dehydration mechanism of Danielsen [1982], convective events introduce tropospheric air into the stratosphere and dehydration results through the sedimentation of ice crystals. The appearance of cirrus, saturation and tropospheric values of ozone mixing ratios correspond well with centres of deep convection seen by the Advanced Very High Resolution Radiometer (AVHRR) and thus suggest that stratosphere-troposphere exchange and dehydration could be taking place. However, the driest air observed by MLS at 68hPa is remote from the region of cirrus formation, convection, coldest temperatures and saturation which is an intriguing puzzle in the understanding of stratosphere-troposphere exchange and the origin of driest air in the stratosphere.

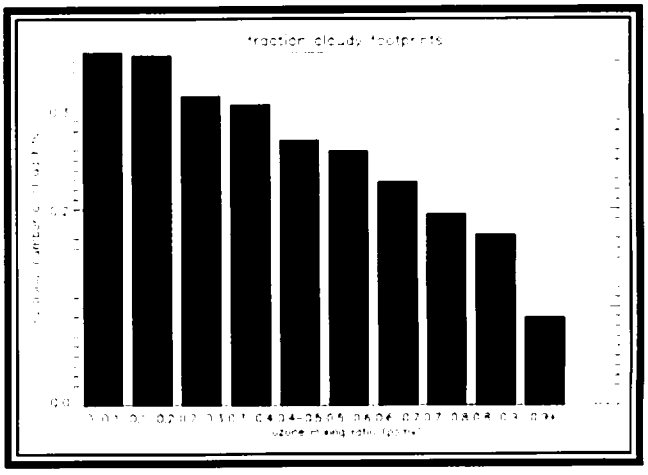

Figure 2
Histogram of the number of CLAES cirrus cloud hits occurring in 10ppmv ozone mixing ratio bins at 68hPa during DJF 1992.

REFERENCES

Danielsen, E.F., A dehydration mechanism for the stratosphere, Geophys. Res. Letts. 9, 605-608, 1982.

Mergenthaler, J.L., A.E. Roche, J.B. Kumer, G.A. Ely, Cryogenic Limb Array Etalon Spectrometer observations of tropical cirrus, J. Geophys. Res., 104, 22183-22194, 1999.

Hannah Clarke and Robert Harwood,
Department of Meteorology, University of Edinburgh.

Making best use of chemical data collected by aircraft in the estimation of photochemical ozone production in the UTLS

Ozone is both an important greenhouse gas and surface pollutant affecting air quality. The ozone distribution in the upper troposphere (UT) is influenced by transport of high ozone concentrations from the lower stratosphere and polluted air from the continental boundary layer. Air with low ozone concentrations from the marine boundary layer can also be transported rapidly into the UT. Ascent from the boundary layer in frontal systems or convection is sufficiently rapid (timescales of a day or less) that ozone precursors, such as nitrogen oxides, can be carried within air masses to the UT where ozone production continues. Therefore, the origin and history of air masses has a strong influence on the ozone distribution and any long-term changes.

In order to make a quantitative estimate of the ozone distribution and possible future changes it is essential to monitor many chemical constituents involved in photochemistry and estimate their distributions. However,

observations are mainly limited to time series from a number of fixed locations and intensive observation periods when aircraft are used to investigate atmospheric composition. It is necessary to use chemical-transport models to estimate the distribution of constituents for the spatial and temporal gaps in the data and to predict future ozone changes.

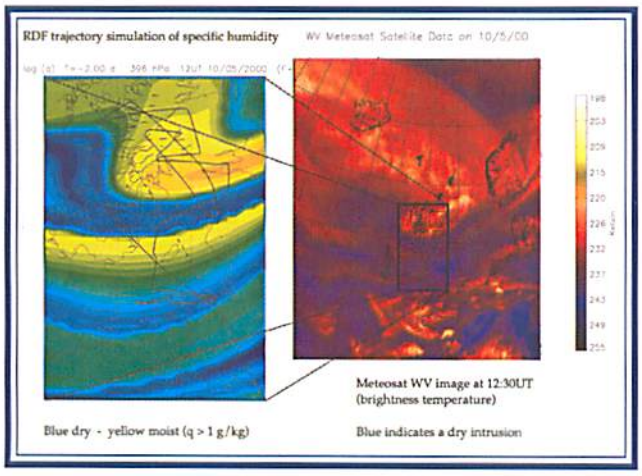

Figure 1a and 1b

The UTLS OZONE project Atmospheric Chemistry and Transport of Ozone (ACTO)* involved a flight campaign using the UK Met. Research flight C-130 to measure ozone, its precursors and indicators of in-situ photochemistry. The second stage of the project involves extensive data analysis to utilise the observations to their best effect. An important tool throughout the project has been Reverse Domain Filling (RDF) trajectory calculations in 3D. In these calculations the trajectories of air masses are traced back in time, by advecting with the wind field from the ECMWF analyses, from points on a regular grid in longitude, latitude and altitude. Temperature and specific humidity at the origin of these trajectories are assigned to the arrival points on the grid. Figure 1a shows the RDF specific humidity field on a horizontal UT surface (400hPa) obtained using two-day back trajectories for the 10th May 2000. Long, narrow filaments running approximately west-east are immediately apparent. These features are only 10s of km wide; much narrower than resolved by the ECMWF analyses themselves (without the trajectory calculations). All the information is contained within the time sequence of the wind fields and the humidity field two days previously, even though these fields have a much larger scale (about 180km). The method works well because the trajectories are 'chaotic', meaning that the origin of neighbouring air masses in the arrival domain can differ greatly. The Meteosat water vapour channel image (Figure 1b) demonstrates that these narrow filaments have been modelled accurately.

RDF forecasts of finescale tracer structures (using ECMWF meteorological forecasts) were used to plan the ACTO flights so that the C-130 aircraft could fly through as many layers with different origins and chemical characteristics as possible. Flight legs in the across-filament direction were used to identify air masses of interest before turning in a pre-determined along-filament direction in order to characterise the air mass with the slower response instruments. Figure 2 shows a height-section through the atmosphere along the dashed line in Figure 1a. The RDF specific humidity reveals the slanting air masses (tracer sheets) that the aircraft flew through (dotted line). The new Met. Research Flight Lyman-alpha water vapour instrument [provided by K. Dewey, MRF] showed remarkable agreement between the structures in the RDF simulation and the observed humidity along the flights (not shown).

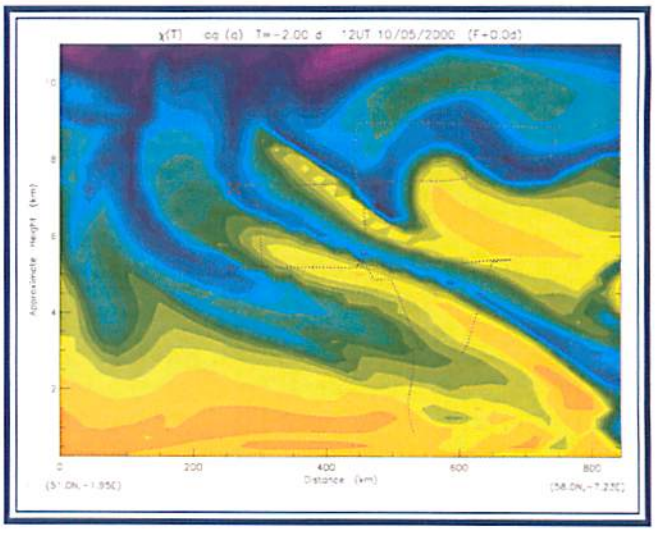

Figure 2

Ozone photochemistry occurs with timescales longer than the stretching timescale for tracer sheets and as a result chemical fields become fairly homogeneous within sheets with large gradients between them. This property is also shared approximately by water vapour. Future work will relate temperature and humidity measurements from flights to the positions in the RDF domain with similar model temperature and humidity. In this way, jumps and narrow peaks in along-flight chemical concentration time series can be related to crossing the edges of air masses seen in the RDF simulation. Chemical concentrations can be inferred throughout the domain from the observations, assuming homogeneity within air masses. Similarly observationally constrained photostationary state models or chemical box models integrated along trajectories (accounting for emissions and deposition) will be used to scale up estimated ozone production rate from the flight path to the whole domain encompassing the UTLS region over the North Atlantic.

John Methven,
Department of Meteorology, University of Reading.

UK Meteorological Office C-130 research aircraft, used during the ACTO campaign. Courtesy of UK Meteorological Office © Crown Copyright.

** = ACTO is a consortium project involving groups from University of East Anglia (UEA), University of Cambridge, Imperial College, University of Leeds, Leicester University and University of Reading.*

Abstracts from

Farnborough Airshow

Helen Rogers was involved with the organisation of an exhibition stand at the Farnborough Airshow - a high-profile bi-annual event. The Airshow took place in July 2000 for one week and is aimed towards trade and the public. The stand was organised by the 'Greener by Design' initiative and Helen Rogers represented EORCU throughout the seven days as an environmental expert. UTLS OZONE's publicity material was on display at the stand and Kathy Law and Rebecca Penkett attended as environmental experts for one day.

Lower stratospheric ozone trends in relation to changes in dynamics

Stratospheric ozone has decreased at Northern mid-latitudes since the beginning of the 1970s and has usually been attributed entirely to anthropogenic ozone deletion, i.e. the increasing concentrations of ozone depleting substances in the stratosphere. However, hemispheric dynamics have also changed since the middle of the 1960s. The influence of changes in tropopause altitude on stratospheric ozone trends was first demonstrated for the ozone series of Hohenpeissenberg (Germany). The North AtlanticOscillation (NAO) index shows a trend from the middle of the 1960s to the middle of the 1990s. We have introduced NAO or the related tropopause pressure, as dynamical proxy in a multiple regression model using total ozone measurements at Arosa (Switzerland). We found that approximately 25% of the winter stratospheric ozone trends during the period from 1970 to 1997 can be explained by changes in hemispheric dynamics, such as NAO or Arctic Oscillation (AO). The influence of changes in hemispheric dynamics is largest in the lowest part of the stratosphere and ozone trends become insignificant up to an altitude of 15km if a dynamical proxy is included in the statistical model (using the ozone sonde record of Payerne, Switzerland). We therefore might conclude, that the ozone trend maximum in the lowest part of the stratosphere reported in the SPARC Asssessment is mainly caused by changes in dynamics. The physical driving mechanism of NAO is presently not known, which precludes the prediction of the dynamical contribution to the future European ozone trends. We strongly recommend using dynamical proxies in future ozone trend analysis to avoid any confusion in the attribution of ozone changes to the success of the Montreal protocol.

Johannes Staehelin, Andrea K. Weiss and Gisela Koch,
Institute for Atmospheric Science, Swiss Federal Institute of Technology, Zürich, Switzerland.
Christoph Appenzeller,
Meteo Swiss, Zürich, Switzerland.

Royal Aeronautical Society Article

UTLS OZONE were invited to write another article for the RAeS. The article entitled, 'The Upper Troposphere/Lower Stratosphere Ozone Programme: contributing to research on aviation impact' featured in The Aeronautical Journal, Vol. 104, No. 1037, July 2000. It also formed part of the display at the Farnborough Air Show together with other UTLS OZONE publicity material. Both Kathy Law and Rebecca Penkett, along with all other authors of the July issue, have been awarded with the 'Hodgson Prize' from the RAeS, which they will receive in July 2001.

Rob MacKenzie (Unversity of Lancaster), John Remedios (University of Leicester) and Don Grainger (Oxford Univeristy) relaxing at the Science Meeting dinner, Leicester, December 2000. Courtesy of Miss R. Penkett.

Long-range transport by frontal systems - impact on ozone and its precursors

This paper discussed case studies and a climatological framework for the transport of pollution from North America to Europe and the role played by upward transport in frontal weather systems.

Anthropogenic emissions from the south-eastern United States have a high probability of getting entrained into warm conveyor belts ahead of cold fronts which lift them into the upper troposphere, where they can be transported across the North Atlantic. North American anthropogenic emissions from these southerly latitudes therefore normally do not reach surface sites in Europe. Case studies of high ozone concentrations in the upper European troposphere, originating from the United States and lifted in warm conveyor belts, show evidence for this mechanism.

In contrast, boreal forest fire emissions take place at higher latitudes and are transported at lower altitudes. Therefore, they have a high probability of reaching European surface sites. It has shown how strongly Carbon Monoxide (CO) emissions from boreal forest fires in Canada affected CO measurements at Mace Head in August 1998 compared to the much weaker influence from both European and North American anthropogenic CO emissions.

Andreas Stohl,
Ludwig-Maximilians-Universität München, Germany.

Web site

The UTLS OZONE web site is aimed at informing browsers about the Programme on topics such as background, funded projects, meetings, publicity and reports/plans etc.. The site was updated in January 2001 to accommodate UTLS OZONE's different users. Browsers can now enter the site from the perspective of a scientist, the public, industry/government or the media. Once a perspective has been chosen, a similar web page to the previous layout appears but with the subjects most relevant to that user at the top of the page, e.g., the Expert Panel (made up from a number UTLS OZONE scientists) is on the top line of the public's page. UTLS OZONE is presently improving links to and from other related sites, which will help to further disseminate information and increase interest in the Expert Panel:

http://utls.nerc.ac.uk/Exp_Pan.html

Additional advertising suggestions for the Expert Panel would be welcome!

Invited Speakers

SCIAMACHY: relevance for UTLS research

The SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) is a passive remote sensing instrument. It is a joint German and Dutch/Belgian contribution to the payload of the European Space Agency (ESA) ENVISAT, which is planned to be launched in October 2001. The Global Ozone Monitoring Experiment (GOME) is a smaller scale version of SCIAMACHY, launched aboard the second European Research Satellite (ERS-2) in April 1995. GOME has now made over five years of successful measurements. GOME-1 successfully demonstrated the SCIAMACHY measurement concept. Improved GOME-2 instruments are part of the three European Organization for the exploitation of meteorological (EUMETSAT)/ESA EUMETSAT Polar System (EPS) Metop missions, planned for the period 2006-2020.

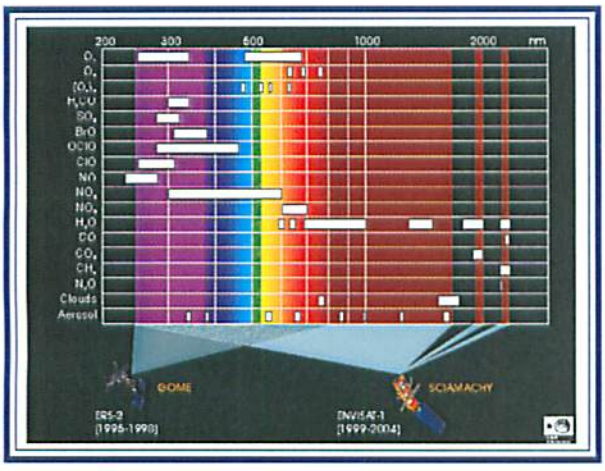

Figure 1

Atmospheric Constituents targeted for SCIAMACHY and GOME.

Both ERS-2 and ENVISAT are designed to fly in sun synchronous polar orbits having equator crossing times in descending node of 10.30am and 10.00am, respectively. SCIAMACHY has eight measurement channels and observes simultaneously the entire backscattered up-welling radiation between 220 and 2380nm: the spectral resolution being approximately 0.2, 0.4, 1.4 and 0.2nm in channels 1 & 2, 3 to 5, 6 and 7 & 8, respectively. The first four channels of SCIAMACHY and those of GOME are equivalent.

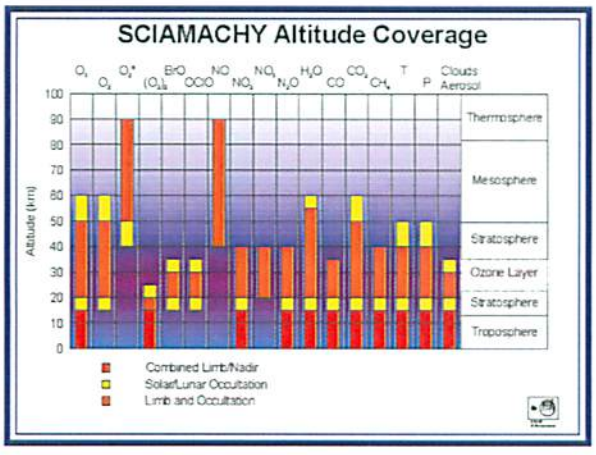

Figure 2

Altitude ranges of the retrievals of atmospheric constituents and parameters to be inverted from SCIAMACHY data.

SCIAMACHY and GOME measure the radiance up-welling from the atmosphere and the extraterrestrial solar irradiance. The difference in the outgoing atmospheric radiance and the incoming solar irradiance yield the atmospheric extinction, which is determined by absorption and scattering in the atmosphere and at the surface of the Earth. The atmospheric constituents (clouds, aerosols and gases), targeted to be retrieved from SCIAMACHY measurements, are shown in Figure 1. The altitude ranges of the retrievals of the targeted atmospheric constituents and parameters is shown in Figure 2. In addition temperature and pressure profiles will be retrieved.

John P. Burrows,
Institute of Environmental Physics and Remote Sensing, University of Bremen, Germany.

Meetings

UTLS OZONE, together with other institutions and funding agencies (including the EC and NASA), jointly sponsored a workshop on NO_x/NO_y in March 2001, Heidelberg, Germany. The workshop aimed at bringing together leading researchers in the field to discuss the latest results and to form an opinion about our current state of knowledge. Kathy Law was a rapporteur during one of the sessions. Klaus Pfeilsticker (IUP-Universität Heidelberg), Kathy Wolfe (Computer Sciences Corporation) and EORCU were involved with the organisation, with Rebecca Penkett's contribution made on behalf of UTLS OZONE and EORCU.

Several scientists funded by UTLS OZONE also presented their results at the European Geophysical Society Meeting, Nice (26th-30th March 2001) in a special session on the Chemistry of the Upper Troposphere and Lower Stratosphere, co-convened by UTLS OZONE.

Atmospheric chemistry and climate change

The Third Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) is in its final stages of preparation before publication this year (2001). The results contributed to the IPCC from the UK Meteorolgical Office's (UKMO) global 3D Lagrangian chemistry model, STOCHEM, point to methane and ozone as the second and third most important greenhouse gases after carbon dioxide in forcing climate change over the next 100 years. The radiative forcing from ozone changes occur largely in the upper troposphere. Global methane emissions are an important driving force as are surface and aviation NO_x emissions. The influence of climate change on tropospheric chemistry is also an important issue from 2050 onwards through changes in the global temperature and water vapour distributions.

Richard G. Derwent, William J. Collins and Cathy E. Johnson,
Climate Research Division, Meteorological Office.
David S. Stevenson,
Department of Meteorology, University of Edinburgh.

Bill Randel (NCAR, Boulder, USA) and Andreas Stohl (Ludwig-Maximilians-Universität München, Germany) relaxing at the Science Meeting dinner, Leicester, December 2000. Courtesy of Miss R. Penkett.

Three-dimensional model studies for the Third European Stratospheric Experiment on Ozone

Our understanding of the processes leading to stratospheric ozone depletion both at high and mid-latitudes has advanced greatly during recent years. Although there remains no doubt that the substantial ozone loss in high-latitude winter and spring is caused by anthropogenic chlorine and bromine emissions, there is still uncertainty about the quantitative extent. For example, there has been a tendency for models to underestimate the loss inferred from observations. Importantly, there are even larger uncertainties about the contribution of anthropogenic halogen trends to the observed ozone trends at mid-latitudes. The Third European Stratospheric Experiment on Ozone (THESEO), an EC funded large field campaign involving a wide range of observations from ground-based, aircraft, balloon-borne and space-borne instruments, which took place in winters 1997/98 and 1998/99 to investigate ozone loss in the Arctic and at mid-latitudes. The campaign was extended during winter 1999/2000, in collaboration with the NASA Sage III Ozone Loss and Validation Experiment (SOLVE) campaign. As part of the UTLS OZONE programme, we have used the SLIMCAT three-dimensional chemical transport model [e.g. Chipperfield, 1999] to perform accompanying modelling studies.

In this article, we focus on two aspects of this project:

1. How much chemical ozone loss occurred in the Arctic stratosphere during recent winter/spring periods? Are the modelled ozone losses consistent with losses derived from observations?

2. Do we understand the role of stratospheric bromine chemistry in determining ozone loss at high and mid-latitudes? Can the model reproduce the observed diurnal, seasonal and latitudinal variations of BrO?

ARCTIC OZONE LOSS DURING WINTER 1999/2000

After the two relatively warm Arctic winters of 1997/98 and 1998/99, the winter of 1999/2000 was characterised by very low temperatures from late November/early December 1999 until March 2000 and a record long period of temperatures below the threshold for possible polar stratospheric cloud (PSC) formation. As a result of these low temperatures, the model calculations showed large levels of chlorine activation which then led to significant catalytic ozone depletion. In addition, the unusually low temperatures caused widespread denitrification - the irreversible removal of nitrogen oxides from the lower stratosphere. This denitrification was observed during THESEO-2000 and crudely reproduced by our model. Denitrification has the potential to increase the overall ozone loss in a given winter because reactive nitrogen that can mediate ozone loss has been removed.

Figure 1 shows a comparison of SLIMCAT model results with ozone sonde observations at the Arctic station at Spitsbergen between November 1999 and April 2000. The model reproduces the observed ozone evolution very well, indicating that the model correctly reproduces the chemical ozone depletion during this period. Comparison with a modelled passive ozone tracer, that has been initialised by the modelled ozone field on 1st December 1999, shows that by the end of March, more than 70% of the ozone has been chemically depleted. The modelled denitrification contributes about 30% to this total loss [Davies et al., 2001].

Although the modelled ozone loss agrees very well in winter 1999/2000, the agreement is not so good with the smaller losses observed in previous years. Winter 1999/2000 was the first year that the model produced significant denitrification in the Arctic and it is possible that the model's apparent underestimation of the ozone loss in previous cold years may be due to our crude treatment of this process which failed to capture observed denitrification. The comparison of the model with historic datasets from previous winters will be an important component of further improving the model and our understanding.

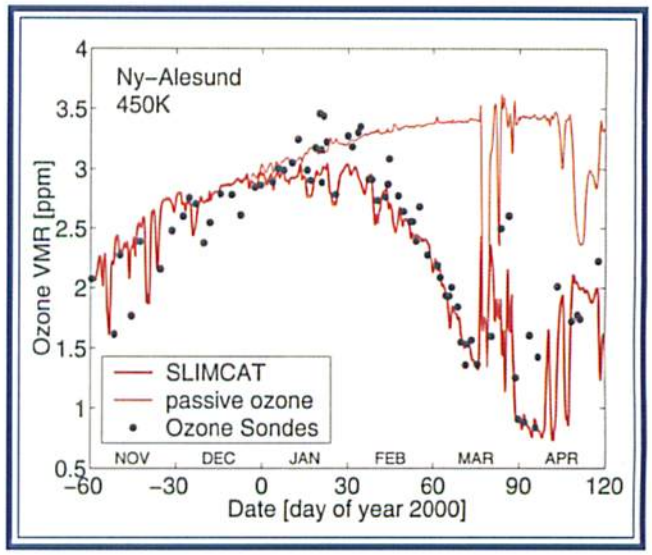

Figure 1

Ozone sonde measurements at Ny-Ålesund, 79°N, compared to the SLIMCAT three-dimensional chemical transport model output at the 450K isentropic level (about 18km altitude). The comparison with the modelled passive ozone tracer shows that by the end of March 2000 more than 2.5ppmv or 70% of the ozone at this level has been chemically depleted. (Figure adapted from Sinnhuber et al. [2000].)

STRATOSPHERIC BROMINE CHEMISTRY

Bromine compounds are believed to play an important role in the destruction of stratospheric ozone, both at high and mid-latitudes. Roughly, 50% of the chemical ozone loss in the Arctic winter can be attributed to catalytic bromine reactions. Despite its importance, stratospheric bromine chemistry is not as well studied as chlorine chemistry. In order to test our current understanding of stratospheric bromine chemistry, we compared ground-based UV-visible measurements of bromine monoxide (BrO), provided by M. Van Roozendael and F. Hendrick, with simulations from the SLIMCAT model. BrO is the only bromine species which is routinely measured in the stratosphere. The measurements were performed at a global network of eleven ground based stations ranging from the Arctic via the mid-latitudes of both hemispheres to the Antarctic. Interpretation of the zenith sky UV-visible measurements is complicated, as the primary measured quantity is BrO slant column densities along the slant path traversed by the scattered sunlight. To allow a direct comparison between observed and modelled BrO, we simulate the measurement geometry by coupling a radiative transfer model to the chemical model to calculate modelled BrO slant column densities.

As an example, Figure 2 shows a comparison of observed and modelled BrO slant column densities for two sites: Harestua at 60°N (top) and Observatoire de Haute Provence at 44°N (bottom). The agreement between the model and

the observations is generally very good. The model reproduces correctly the seasonal and latitudinal variations of both the absolute amount of the BrO observations as well as their diurnal cycle, indicating that our understanding of the BrO related bromine chemistry is basically correct. The agreement shown in Figure 2 is better than that shown in many previous model/observations comparisons. Generally this improvement is due to a number of incremental factors including improved laboratory data for some key photochemical processes, improved algorithms for the retrieval of BrO and improved modelling techniques allowing the direct comparison of measured quantities. Moreover Figure 2 shows how the comparison of measurements and model can help to assess the internal consistency of the BrO time series at one station and the consistency between the BrO measurements at different stations.

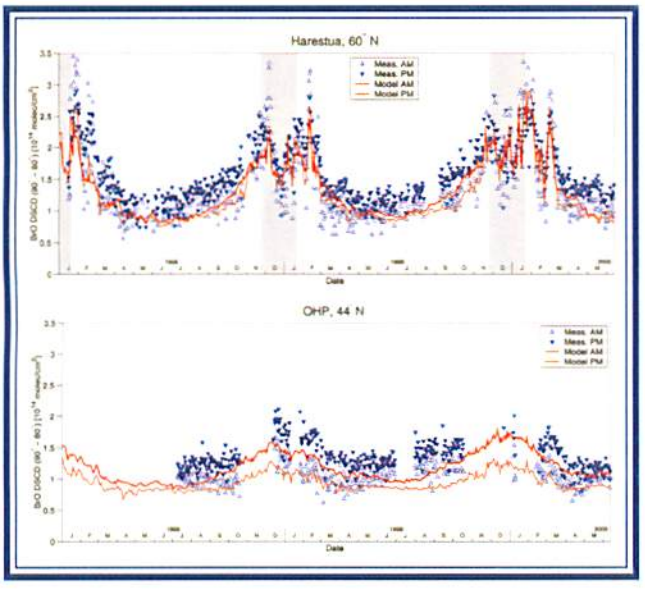

Figure 2a & 2b

Comparison of observed and modelled BrO differential slant column densities for Harestua, 60°N, (top) and Observatoire de Haute-Provence (OHP), 44°N, (bottom).

Based on the results from this comparison with BrO observations, indicating a good understanding of BrO chemistry, we are currently using the model to assess the impact of the changes in stratospheric bromine chemistry on ozone depletion.

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Davies, S., et al., Modelling the effect of denitrification on Arctic ozone depletion during winter 1999/2000, submitted to J. Geophys. Res., 2001.

Sinnhuber, B.-M., et al., Large loss of total ozone during the Arctic winter of 1999/2000, Geophys. Res. Lett., 27, 3473-3476, 2000.

ACKNOWLEDGEMENTS

We thank Michel Van Roozendael and Francois Hendrick from the Belgian Institute for Space Aeronomy for providing the BrO data and helpful discussions.

Björn-Martin Sinnhuber and Martyn P. Chipperfield,
School of the Environment, University of Leeds.

The Aberystwyth Egrett Experiment*: Mixing in the Tropopause Region

During May and June 2000, a diverse group of scientists and engineers (inside front cover) conducted an airborne measurement campaign to study the dynamics of mixing in the tropopause region. This experiment made use of a very unique aircraft called the Egrett. Flying at heights of up to 14km and at relatively low airspeed, a very wide spectrum of processes could be observed, from large scale lateral filamentation, to small scale gravity waves and turbulence.

The Egrett was equipped with an advanced turbulence measurement system and instruments for sampling ozone, water vapour, methane and CFCs. Most of the flights were directed above Aberystwyth in order to combine results with ground based measurements from the Mesosphere, Stratosphere, Troposphere (MST) radar, an ozone lidar and balloons. Over a 6-week period, the Egrett was based at Boscombe Down in Wiltshire and spent 90 hours in the air above Britain and Ireland in 16 separate flights.

The flight time was equally divided between two main scientific objectives.

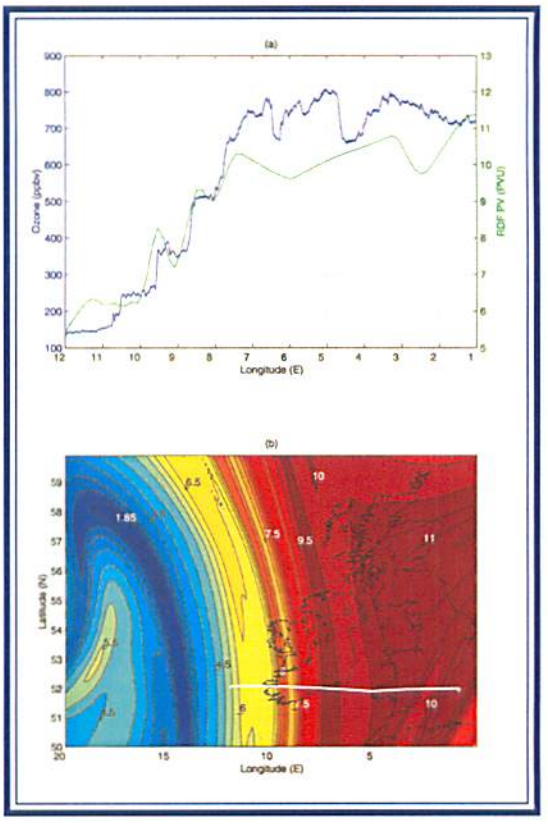


Figure 1a & 1b

(a) Ozone mixing ratios measured on the Egrett at a height of 14km as it flew eastward above Ireland and Wales. The potential vorticity (PV) computed from 2-day Reverse Domain Filling (RDF) back trajectories is also shown.
(b) Contour plot of RDF PV on the 385K isentropic surface. The white line shows the Egrett flight path.

OBJECTIVES

1. To study the lateral mixing between air from different latitudes. For example, Figure 1a shows the measured ozone abundance as the Egrett flew eastward through the edge of a tropical airmass that was displaced poleward by a planetary (Rossby) wave. The ozone content of the air increased from 150ppbv in the air of tropical origin to 800ppbv in the

** = This project is a consortium of scientists from the University of Wales, Aberystwyth and the University of Cambridge.*

mid-latitude air. The fascinating aspect of this observation is that the ozone increased in steps. These steps are interpreted as being a manifestation of filaments that developed along the interface and this is the initial stage of mixing beween the two different types of air. This filamentation is also seen in the distribution of potential vorticity that is calculated using reverse domain filled back trajectories (Figure 1b).

2. To study the vertical mixing caused by gravity waves and turbulence. These flights consisted of vertically stacked flight legs over the mountains of Wales. Figure 2 shows measurements in the region of maximum shear at the top of the jet stream on the 6th June 2000. The most intense turbulence observed during the campaign occurred in the second half of this flight leg, with gusts in the vertical wind of up to 3m/sec. These wind measurements have a horizontal resolution of about 2 meters. Associated with this patch of turbulence are striking changes in ozone and water vapour. The ozone mixing ratio dropped by 20ppbv while water vapour increased by 1.7ppmv. Further analysis has shown that these anomalies can be accounted for by the turbulent mixing and gravity wave motions.

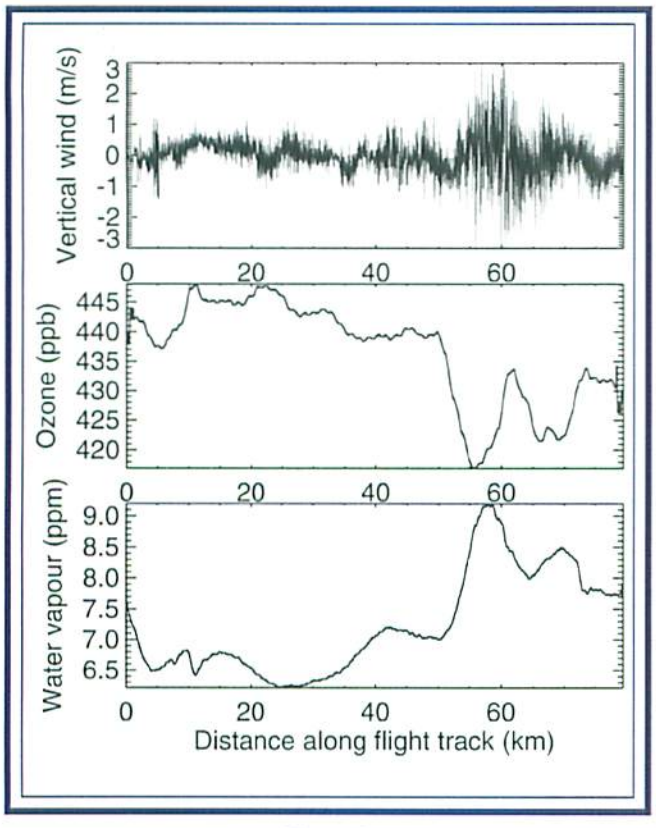


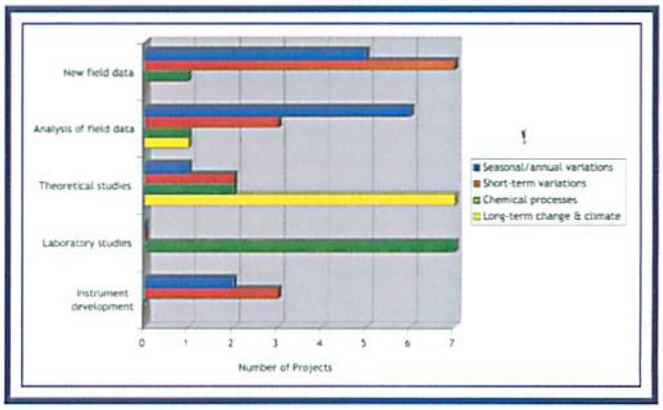
Figure 2

Egrett measurements at a height of 11.4km along a horizontal flight leg on the 6th June 2000.

The two examples shown here demonstrate mixing processes at opposite ends of the spectrum, occurring essentially within the same meteorological setting (24 hours apart). At intermediate scales there were striking measurements of gravity wave breaking. The most challenging goal of the project now is to develop an understanding of mixing that involves all the processes, from planetary wave breaking, through gravity wave breaking and jet stream shear, to turbulence.

Jim Whiteway,
Physics Department, University of Wales, Aberystwyth.

Research Projects and Data Management



The number of projects funded by UTLS OZONE within each research theme and according to the type of activity being used to obtain results.

Data collected and produced as part of UTLS OZONE funded projects are being archived at the British Atmospheric Data Centre (BADC). Initially access to this data is restricted to UTLS OZONE funded researchers only. This gives those involved in collecting and producing the data a chance to publish their new results. The data will be made available more widely after two years. The data archived so far on the BADC includes Hercules C-130 data from the Dynamics and Chemistry in Frontal Zones (DCFZ) and ACTO campaigns, ozone sonde and ozone LIDAR profiles from Aberystwyth, and radar images from the Chilbolton radar. The BADC is also responsible for providing third party data support to UTLS OZONE projects. This has included providing maps of ECMWF forecasts via the web for use in campaign planning and the development of a web interface to a trajectory model. Recently the BADC has started to acquire global and mesoscale assimilation products from the UK Met. Office. The high-resolution mesoscale data (approximately 12km in the horizontal) will provide an invaluable resource for data analysis. For more information on data held at the BADC visit:

http://www.badc.rl.ac.uk/data/utls-ozone/

or contact: badc@rl.ac.uk

Jamie Kettleborough,
Rutherford Appleton Laboratory.

The jet-engined BAE 146-300 aircraft "G-LUXE". is the new NERC-Meteorological Research Flight aircraft taking over from the C-130. After modifications it will have a range of 2000 neutical miles, a celling of 35000 feet, a maximum endurance of six hours, and will carry two crew and up to eighteen scientists. Photo courtesy of BAE Systems. see: http://personalpages.umist.ac.uk/staff/Bob.Wells/aircraft/

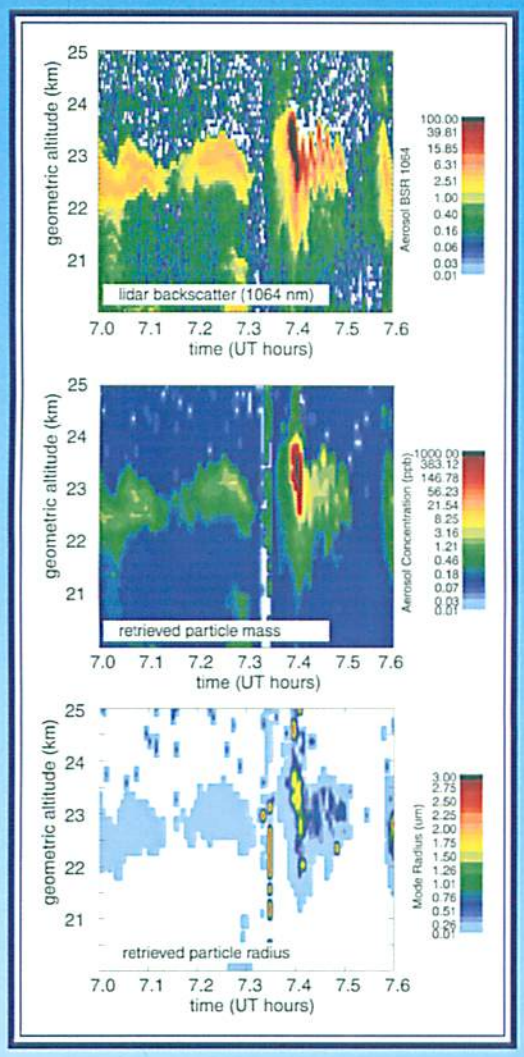
Principal Investigator *	Round 4 Funded Projects
Allen (RAL)	The role of ozone in determining the response of the coupled ocean-atmosphere climate system to solar forcing
Carslaw (Leeds)	Retrieval of polar stratospheric cloud microphysical properties from multi-wavelength satellite extinction and lidar measurements
Cox (Cambridge)	Oxygenated organics in the tropopause region - kinetics and mechanism of formation
Ford (UCL)	Freezing processes in aircraft contrails and atmospheric aerosol
Grainger (Oxford)	Satellite observation of aviation impacts on the UTLS
Gray (RAL)	Interannual variability associated with the QBO and its impact on the UTLS region
Harwood (Edinburgh)	Chemistry-climate modelling of the present and future UTLS
Horn (York)	Simultaneous mechanistic and kinetic studies of heterogeneous atmospheric chemistry on soot
MacKenzie (Lancaster)	Cirrus and aerosol properties in and around the south European air traffic corridor
Penkett (UEA)	Use of CARIBIC data to determine the source of trace gases and aerosols in the UTLS region

** Note that, due to space restrictions, only the first named principal investigator is given.*

For a complete list of UTLS OZONE funded projects, please go to:
http://utls.nerc.ac.uk/projects/project.html

Selection of photos from the Aviation Impact Studies Workshop Dinner, October 2000, RAeS. Courtesy of participants.

Contact Information



Lidar observations of a mountain wave-induced polar stratospheric cloud together with the derived cloud microphysical properties. The cloud was observed by the Langley/GSFC aerosol lidar over East Greenland during a transit flight of the DC-8 as part of the SOLVE/THESEO-2000 campaign. Three different clouds are apparent (liquid aerosol, ice, and nitric acid hydrate). The microphysical properties (lower two panels) have been retrieved using a new technique developed by Rongming Hu and Ken Carslaw, University of Leeds, as part of their UTLS OZONE project. Courtesy of Dr K. Carslaw.

Photo courtesy of NASA.

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