

# ACTIVE: Aerosol and chemical transport in tropical convection

## 1. Overall scientific case

### 1.1 Introduction

The goal of this proposal is to answer a number of key questions about the upper tropical troposphere, a critical research area for climate change as well as the evolution of stratospheric ozone. Its focus is on the role that deep convection in the tropics plays in transporting aerosol and chemical species from the planetary boundary layer to the upper troposphere. This issue is crucial for climate science (since the aerosol serve as cloud condensation nuclei) and for the overall composition of the tropical tropopause layer (TTL) – the layer of the tropical atmosphere between about 13 and 17 km altitude. This in turn determines the composition of the global stratosphere. We aim to make direct measurements of a range of aerosol and chemical species in the low-level inflow and the high-level outflow of tropical thunderstorms, relating these to model simulations of the storms and the wider environment in which they are embedded.

The proposal is based on aircraft campaigns at Darwin, Australia (12°S, 131°E), using the new NERC community aircraft for low levels and the Australian Egrett for the high-altitude outflow. Darwin is the ideal location for the experiment, firstly because it has excellent infrastructure for the aircraft; secondly because of the existing ground-based suite of radars, radiometers and meteorological instruments; and thirdly because of the predictable nature of deep convection in this region. The present proposal builds on the experience already gained by UWA and UMIST at Darwin during the EMERALD-II campaign to study the nature of the cirrus outflow from tropical storms. The new campaign will take place in the period November 2005-February 2006, as part of two international experiments: the joint Australian/NASA/ARM TWP-ICE campaign at Darwin in early 2006 to study cloud and rain characteristics in the Australian monsoon, and the EU SCOUT-O3 campaign to study the composition of the TTL (using the Russian Geophysica and German Falcon aircraft, probably from Darwin in late 2005). The synergy between the proposed measurements and those from our international partners will generate a uniquely powerful dataset for constraining and developing models of vertical transport in the tropics and the structure of the TTL.

### 1.2 The science problem

The thermal structure of the tropical troposphere is determined by radiative-convective equilibrium up to 12-14 km, which is the level of outflow of most of the convective storms (Folkins 2002). Above this, the temperature continues to drop to a cold point around 17 km, but the frequency of convective penetration decreases with height and the atmosphere approaches radiative balance (Highwood and Hoskins 1998, Thuburn and Craig 2002). It therefore behaves as the base of the Brewer-Dobson circulation, determining the eventual composition of the global stratosphere. Below the TTL convective processes dominate the vertical transport of chemical species. The vertical transport is fast and there are potential cloud-chemical processing effects. In the upper part of the TTL non-convective processes dominate and the corresponding vertical transport is slow. A key quantitative uncertainty in the TTL is the partition, as a function of height, horizontal location and season, between convective and non-convective transport.

Air transported by deep convection to the upper troposphere will have distinctive properties, compared with air transported by large-scale ascent. For a start, short-lived chemicals of land, marine or anthropogenic origin can be deposited virtually unchanged at high altitudes. The source gases for halogen radicals for instance (particularly bromine and iodine) could be transported in this way, affecting ozone concentrations in the TTL and, subsequently, the lower stratosphere (see Chapter 2 of WMO, 2002). The stratospheric impact of the halogen compounds depends on the interplay between the transport and the chemistry in the TTL, both of which are poorly understood. This interplay determines the degree of transformation that the source gases can undergo in the TTL, and how much bromine and iodine actually enters the stratosphere. Conversely, observations of some short-lived halogen compounds can reveal the origin of air transported by deep convection: CH<sub>3</sub>I is present in marine air, and so its presence in the TTL indicates recent marine convection (Cohan et al., 1999).

Lightning in deep cumulonimbi is a potent source of NO<sub>x</sub> radicals, and possibly also of ozone; the EMERALD-II campaign observed elevated ozone in anvils (see below) the source of which is currently uncertain. In any case, elevated NO<sub>x</sub> with hydrocarbons (VOCs) of ground-level origin provide a photochemical source of ozone in the upper troposphere. As ozone is an effective greenhouse gas in this region a better understanding of its chemistry is very important for climate modelling.

Crucial to our ability to forecast future climate change is a better representation of cirrus clouds in climate models. This in turn means understanding the density and nature of the cloud particles that are found in deep convective anvils, since deep convection is the major source of cirrus cloud in the tropics. This was the main thrust of EMERALD-II (see below). However, in order to be able to calculate the crystal number concentration and sizes in the cirrus outflow from a thunderstorm, it is necessary to know the aerosol population, since it is from this population that the cloud condensation nuclei are drawn. There have been few measurements to date of the aerosol population associated with deep tropical thunderstorms, and models of convective transport of aerosols and chemicals are largely untested; although comprehensive studies of the microphysics of cirrus anvils were performed as part of the Cirrus Regional Study of Tropical Anvils and Cirrus Layers – Florida area Cirrus Experiment (CRYSTAL-FACE) during July 2002, the work proposed here will focus on the role of deep tropical convection on the aerosol and composition of the UTLS region.

The life cycle of aerosol in the atmosphere remains uncertain. One picture involves the formation of fresh ultrafine particles in the outflow of rapidly uplifted air. The uplift causes significant aerosol loss through precipitation, but it also transports non-soluble gaseous precursors that can subsequently be oxidised to nucleate new ultrafine particles (Raes et al., 2000). The lack of surface area, cold temperatures and increased ion abundance close to the tropopause all favour new particle formation and the long residence times in the TTL allows for growth to several tens of nanometres where particles may participate in optical and cloud-forming processes. New particle formation in the TTL may well provide sufficient particle number to supply the lower stratospheric burden (Brock et al 1995). Twohy et al (2002) studied the outflow from anvils of a mesoscale cluster over the mid-western USA to investigate new particle formation in the outflow region. They found CO concentrations rising to 130 ppbv within the outflow, compared with 100 ppbv outside and CN concentrations increasing to  $4 \times 10^4 \text{ cm}^{-3}$  within the outflow, more than an order of magnitude greater than outside it. Further, NO concentrations increased to 1500 pptv within the outflow compared to near zero outside. No measurements were made in the inflow region of the cloud but the study clearly demonstrated the effectiveness of deep convective clouds in transporting material (gases and particles) into the tropopause region followed by detrainment from the cloud by cirrus anvils. The study also presented strong evidence for the formation and growth of new sulphate particles in this region of the atmosphere.

It is well established that the black carbon content of atmospheric aerosols provides an important absorber of visible radiation in the atmosphere leading to local warming. The black carbon budget in the UTLS remains very uncertain, and whilst aircraft are a significant source, predictions show they do not provide the main source in the tropics where it is likely that convective transport of smoke from biomass fires provides a considerable source (Hendricks et al 2004). Recent measurements, with a single particle soot photometer, show considerably more black carbon in the Arctic stratosphere than has previously been measured (Baumgardner et al 2004). If these are correct the budgets must be substantially corrected and their role in warming the lower stratosphere will be significant. There are no such measurements in the tropics and this should be seen as a priority that ACTIVE will seek to address. If black carbon is mixed in the same particles as sulphate aerosol or dust, the sign of the resulting radiative forcing can be reversed, since the particles absorb as well as scatter. This points to a major difference between fresh and aged soot. Furthermore, black carbon can act as an efficient ice nucleus, affecting the ice crystal number and also its optical properties. Its distribution across the aerosol population will have an important bearing on the number of available IN in the TTL region. It has to date not been possible to make such measurements since the methodology and instrumentation has not existed. ACTIVE will make single-particle black carbon measurements to provide a unique data set in the TTL.

As well as uncertainties in composition, there are significant inadequacies in current large-scale chemical transport models and chemical-climate models in the TTL. Firstly, the transition from convective transport dominating to non-convective transport dominating takes place over two or three km (i.e. only two or three grid points even in models with relatively high vertical resolution). Secondly, the velocity fields from meteorological datasets that are used to drive chemical transport models, both grid-based models and trajectory models, are likely to be poor in this region, in part because of the subtle mix of convective and non-convective processes. Slow vertical transport in non-convecting parts of the TTL is liable to be swamped by the effects of poor resolution and poor representation of these processes. Recent experience in DAMTP with trajectory calculations in the TTL based on ERA-40 3-dimensional velocity fields shows unrealistically large heating rates on many trajectories. Chemical measurements of the type proposed here are needed to deduce with better accuracy the effect of large-scale transport, and disentangle this from convective transport.

Our overall picture of the TTL is almost certainly too simple - a consequence, in part of the small number of quality measurements in the region. Tuck et al. (2004) have recently pointed to the dearth of measurements of the horizontal structure of the TTL and to the inadequacy of conceptual pictures based on simple 1-dimensional

(ascent/descent) treatments, confirmed by recent balloon measurements of tracers in the TTL by the Cambridge group. Based on flights in the northern tropics and sub-tropics, these point to a rich structure and variability. Tracer measurements from Darwin during 2005/2006 will allow the impact of the two different regimes on the structure of the TTL to be assessed. These will represent an important new data set to complement the northern hemispheric data reported by Tuck et al. (2004).

### 1.3 What did we learn from EMERALD-II?

The EMERALD-II experiment was conducted in Darwin by Aberystwyth, UMIST and Imperial College in November-December 2002 using the Airborne Research Australia King Air and Egrett aircraft. Whilst the focus of EMERALD-II was on the effects of the anvils on the radiation field, the experiment provided significant insight into the microphysics of large tropical storms, as well as proving the suitability of Darwin for aircraft studies of this type. No aerosol measurements were made during EMERALD-II; however, cloud-resolving model results have shown that the microphysics of the storms are highly sensitive to the aerosol entering the base and mid levels of the cloud. Hence, it appears that knowledge of the aerosol entering the cloud is vital to a quantitative prediction of cloud microphysical and dynamical behaviour, precipitation development and net radiative forcing. These key issues have provided a further stimulus for the work proposed here.

During EMERALD-II, the two aircraft were based in Darwin and flew 13 sorties towards the Tiwi Islands (see next section). A cloud lidar was flown on the ARA King Air aircraft, measuring the backscatter and polarisation of the cirrus anvil through which the Egrett was flying; the two aircraft followed the same ground path simultaneously several kilometres apart vertically. An example is shown in fig 1 (top panel) of the lidar backscatter with the Egrett's path superimposed as a black line. The surrounding panels show ice crystals observed with the cloud particle imager at the points denoted by the arrows. The P.I. of EMERALD-II, Dr Jim Whiteway, is now a professor at York University, Toronto, and is a collaborator in the present experiment. He is providing a lidar instrument for the Egrett and contributing 40% of the Egrett flight costs.

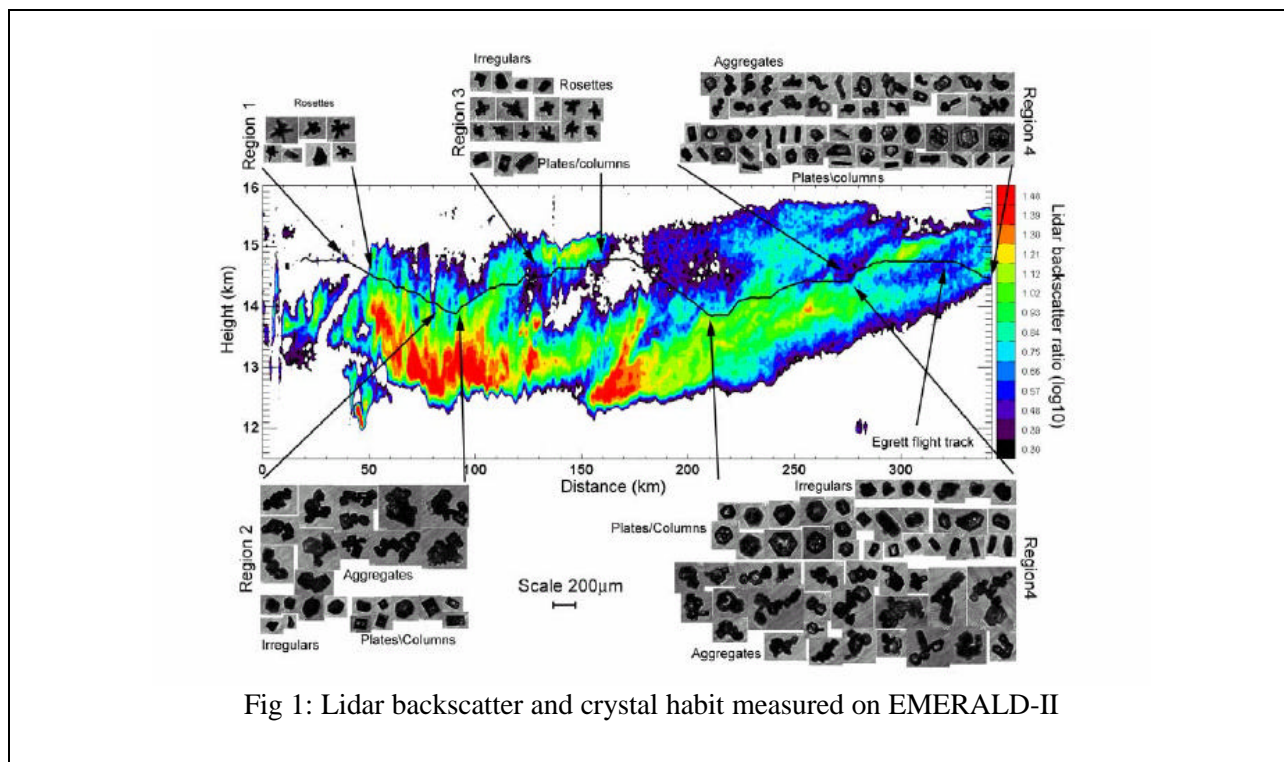


Fig 1: Lidar backscatter and crystal habit measured on EMERALD-II

Analysis of the EMERALD-II data set has revealed differences in the cirrus outflow from the thunderstorms between consecutive days (e.g. in ice number concentration, ice water content, size and trends in the vertical). Analysis of MODIS satellite data shows that aerosol concentrations may be responsible for these differences as it was found that bush fires were affecting the aerosol optical depth. Bush fires are a regular occurrence in the region during November and are significant sources of atmospheric particulates. Large Eddy Modelling (LEM) of EMERALD-II case studies has successfully simulated the storms and predict:

- a) the CCN concentration in the inflow determines the droplet number concentration in the storm. An intermediate value of droplet number concentration causes 'optimal' development of the storm, leading to rapid accretion of cloud water by rain, and glaciation via raindrop freezing/capture. Ice number concentration in the

anvil is correlated with the droplet number concentration, and therefore the CCN input, due to the preponderance of homogeneous freezing.

- b) as the number of ice-forming nuclei (IN) entering it increases, the storm increases in intensity, producing more condensed mass and precipitation. At low IN concentrations, ice number concentration in the anvil is dominated by homogeneous nucleation occurring at  $-38^{\circ}\text{C}$ , whereas for very high values of IN the Bergeron-Findeison mechanism is effective lower down in the cloud.

The overall conclusions from these modelling studies and their comparison with the EMERALD-II data are that, whilst it is possible to simulate the development of the storm, aerosol measurements must be provided as input parameters to properly constrain the model and hence develop its predictive capability. Aerosols, both as IN and CCN, markedly affect the development of the cloud both dynamically and microphysically. These will in turn alter the ability of the cloud to transport and detrain material into the TTL. The data set from EMERALD-II could not be used to provide such tests as no aerosol measurements were made. This proposal aims to use the BAe146 aircraft to provide a very comprehensive suite of physical and chemical measurements of aerosol particles entering the cloud, coupled to a detailed in situ and remotely sensed picture of the cloud microphysics in the anvil region, that will provide a sound test of model performance and prediction.

A surprising result from EMERALD-II was the high concentrations of ozone and water vapour measured in the outflow. The enhanced ozone (up to 150 ppbv) was highly correlated with very high humidity (up to 200% RH wrt. ice) and also with sampling of very large chain-aggregate crystals that would have been formed lower in the convection. Such high humidity suggests that very little aerosol is getting out of the storm, but this could not be confirmed with the EMERALD-II measurements. In the present experiment we will measure a suite of chemical species, as well as aerosol, which will allow this issue to be resolved. Remote sensing of ozone and water vapour by lidar will reveal any instrumental artefacts in the in-situ data.

#### *1.4 Why Darwin?*

Darwin is the ideal location for an experiment of this type, for a number of reasons; that is why the TWP-ICE consortium is planning their experiment there for early 2006 (and why SCOUT-O3 may deploy to Darwin in 2005/6). Firstly there is Darwin's location, giving access to two distinct convection types. From about mid-December to mid-February Darwin is affected by a strong monsoon circulation; the major global monsoon heat source is located over northern Australia in January (Manton and McBride 1992). This results in widespread convective activity over the Darwin area, forming mesoscale complexes typical of convection across the Pacific warm pool region (Toracinta et al 2002). Such conditions occur around 45% of the time in the period November – March (Jakob and Tselioudis 2003) and are generally associated with westerly winds. Measurements in these conditions will allow the contribution of the general warm pool convection to the TTL composition to be determined.

In the pre-monsoon period (mid-November – mid-December) and during monsoon breaks, convection is dominated by single isolated storms. Spectacular examples occur over the Tiwi Islands north of Darwin, due to convergence of sea breezes over the islands (Carbone et al, 2000). These are the Hector storms, which occur predictably over the islands at about the same time each day. These giant thunderstorms, which are deeper and more electrically active than the monsoon storms, were the subject of EMERALD-II. They reach (and penetrate) the tropopause (at 17 km) thus directly injecting air into the TTL and TLS (tropical lower stratosphere). Their predictability and isolation also make them excellent laboratories for comparing the aerosol input and output since the task of modelling them is not so formidable as for extensive convection, as the success of the EMERALD-II LEM modelling testifies.

Darwin also provides the best facilities available to support the experiment. The Australian Bureau of Meteorology (BoM) is keen to foster international research collaboration and Darwin airport provides an excellent base for aircraft operations, as the evidence of EMERALD-II shows. The BoM operates two weather radar systems within 25 km of each other around Darwin, one fully polarimetric and one an operational Doppler. These radars provide real-time images of the convective storms to guide aircraft operations, and their measurements of the 3-D wind and precipitation field in the storms serve as a key constraint for LEM modelling. The BoM also operates 920 and 50 MHz wind profilers about 5 km from Darwin. In addition, Darwin is a fully-instrumented Atmospheric Radiation Monitoring (ARM) site, with comprehensive radiation measurements, micropulse lidar and 35 GHz cloud radar. Radiosondes are launched from the airport, and for January-February 2006 will be launched at 3-hourly intervals from Darwin and a further five stations located around it (including the Tiwi islands and a ship to the west of Darwin).

### 1.5 Why two campaigns?

Two campaigns are planned (before and after Christmas) for two reasons:

- a) the very different meteorology in the two periods. The pre-monsoon period, as mentioned above, is characterised by isolated, giant thunderstorms reaching the tropopause. These provide the opportunity to study the direct injection of material from low level throughout the TTL. Progress in EMERALD-II in modelling these thunderstorms (see above) also demonstrates that we can relate measurements made in the inflow and outflow with some confidence, thus making progress on the basic scientific objectives of aerosol modification. By contrast, the monsoon period has more widespread, less penetrating convection more typical of the Pacific warm pool. Studies during this period will be more readily generalised to the global scale.
- b) The opportunity to participate in wider international experiments. In January and February the TWP-ICE experiment will provide an enhanced infrastructure and a wealth of cloud measurements from ground-based and aircraft platforms. Our aerosol and chemical measurements will complement TWP-ICE, and benefit in turn from their measurements. The SCOUT-O3 experiment will be using the Russian Geophysica and DLR Falcon aircraft to study the TTL in late 2005/ early 2006 and there is a good chance that it will be based in Darwin where it would be able to reach the top of the Hector storms. Both experiments are described in more detail below.

In summary, this is a unique opportunity for UK scientists to address some extremely important scientific questions in collaboration with two large scientific consortia. Our experience and expertise with EMERALD-II, together with the exciting possibilities using the new BAe146 aircraft, means we can make a powerful contribution to these experiments (with aerosol and chemical measurements) and gain a dataset that will provide exacting constraints on a range of models.

### 1.6 Why a consortium?

Consortium grants are intended to support focussed, co-ordinated, collaborative research into specific issues that cannot be addressed through other NERC funding modes. The experiment we propose is very ambitious. It requires several aircraft with a range of specialised instrumentation, and a range of modelling approaches to interpret the data. No one UK university has the necessary expertise to undertake this experiment, but the four universities in this consortium all have wide experience of field campaigns and data interpretation, with modelling capabilities ranging from cloud microscale processes to global transport scales, and their scientific expertise complements and strengthens one another. The project is not suitable for a standard grant because of the cost and the very close co-ordination that will be needed, particularly in the measurement campaigns.

Further justification for a consortium approach comes from the number of international partners. Key instrumentation for the Egrett will be provided by DLR, Germany, (ozone and water vapour) and FZJ (NO, NO<sub>2</sub>) who are particularly interested in the ozone/NO<sub>x</sub> produced by the storm. The CAPS aerosol instrument will be provided by Prof. Andrew Heymsfield, NCAR, USA, who is a world-leading authority on cirrus cloud. The cost of flight hours on the Egrett will be shared by Prof. Jim Whiteway of York University, Canada, who was the PI on EMERALD-II and has extensive experience of interpreting aircraft data. ARA Australia, will provide the basic meteorological measurements on the Egrett, and the Australian Bureau of Meteorology (BoM) will provide ground facilities and supporting measurements at Darwin. The experiment proposed here forms part of two major international campaigns (described below), again requiring a high degree of co-ordination best accomplished through the consortium mechanism.

## 2. Specific Scientific Objectives

The overall aim of this proposal is to determine the impact of deep convection on the composition of the TTL and its relative importance compared with large-scale ascent.

- i. To relate measurements of aerosol and chemical species in the outflow of deep convection, and in the surrounding TTL, to low-level sources.
- ii. To determine how deep convection modifies the aerosol population reaching the outflow, and thus evaluate its impact on cirrus nucleation in the TTL.
- iii. To compare the concentration of aerosol and chemical tracers (with a range of lifetimes and origins) in the outflow with that in the background TTL, and therefore determine the contribution of convection to the composition of the TTL over Darwin.
- iv. To use latitudinal surveys of tracers, together with high-resolution global models, to determine the contribution of large-scale transport to the composition of the TTL

- v. To compare the effects of isolated very deep convection with that of widespread but less penetrative convection on the composition of the TTL.
- vi. To determine the contribution of deep convection to the NO<sub>x</sub> and O<sub>3</sub> budget in the TTL
- vii. To measure how much black carbon reaches the outflow regions of the storms.

### 3. The international dimension

#### 3.1 Tropical Warm Pool – International Cloud Experiment (TWP-ICE)

(<http://science.arm.gov/~mather/darwiniop/>)

The impact of oceanic convection on its environment and the relationship between the characteristics of the convection and the resulting cirrus characteristics is still not understood. An intense airborne measurement campaign combined with an extensive network of ground-based observations is being planned for the region near Darwin during January-February, 2006 to address these questions. TWP-ICE will be the first field program in the tropics that attempts to describe the evolution of tropical convection, including the large scale heat, moisture, and momentum budgets, while at the same time obtaining detailed observations of cloud properties and the impact of the clouds on the environment. The experiment is a collaboration between the US DOE ARM project, the Australian Bureau of Meteorology (BoM), NASA and several United States, Australian, Canadian, Japanese and European Universities.

Observations of cloud properties will be provided by an extensive set of in situ and remote sensing instruments. The Darwin ARM site will provide continuous measurements of cloud properties through remote sensing retrievals. A second set of instruments will be deployed on a ship in the Timor Sea, approximately 100 km northwest of Darwin. Added to these surface-based observations will be several research aircraft including the NASA WB-57 and the DOE Proteus, as well as three aircraft provided by Airborne Research Australia (ARA): the Egrett, King Air and Dimona. These aircraft fulfill a variety of observational requirements to address the science goals (see 4.1). Specific goals of TWP-ICE are:

- Make detailed measurements of the cirrus microphysics and how they relate to storm intensity and proximity (spatial and temporal) to the parent convection.
- Verification of remotely sensed microphysical measurements.
- Provide data sets for forcing cloud resolving and single column models that will attempt to simulate the observed characteristics and impacts.
- Document the evolution of oceanic convective clouds from the early convection phase through to the remnant cirrus with particular emphasis on their microphysics.
- Measure the dynamical and radiative impacts of the cloud systems.
- Characterize the environment in which the cloud systems occur.
- Document the evolution of the convective boundary layer throughout the diurnal cycle and through the lifecycle of convective systems.

#### 3.2 SCOUT-O3

SCOUT-O3 is a 5 year EU integrated project which started in May 2004, with Prof Pyle and Dr Harris as its coordinators. Tropical activities are a major component within SCOUT-O3 and include field campaigns, studies of past and future ground and satellite data, and modelling. The main field campaigns will involve aircraft and balloons. Several measurement phases are being planned. The largest phase will involve the high-flying M55 Geophysica and the DLR Falcon looking at transport of air from the TTL into the stratosphere and is due to take place in late 2005/early 2006. The location will be finally decided in autumn 2004 after further planning studies have taken place. The main options are the Indonesian maritime continent (with the planes most likely based at Darwin) and West Africa. If the M55 and Falcon are based in Darwin in either late 2005 or early 2006, the Egrett and BAe-146 activities proposed here will be closely coordinated with them. The M55 is capable of flying up to 21 km and will carry a range of chemical and particle instruments. It is very well suited to observe the influence of convective outflow on the TTL at higher altitudes than can be measured by the Egrett. The Falcon will carry an ozone and water vapour lidar. It will act primarily as a pathfinder for the M55 guiding it to regions of interest, but it can also perform coordinated flights with the BAe-146.

#### 3.3 Synergy with SCOUT-O3 and TWP-ICE

It is important to see the current proposal in the context of SCOUT-O3 and TWP-ICE. Basically, TWP-ICE is focusing on cirrus while we are focusing on aerosol and chemical measurements (although cloud particle measurements will of course be made by the Egrett and TWP-ICE aircraft will make aerosol measurements). The

Proteus aircraft flies higher than the Egrett and has a flight endurance >10 hours. As well as radiometers, it carries in-situ sampling instruments: a CAPS (as on the Egrett) for cloud and aerosol, frost-point hygrometers, cloud lidar and radar and ice particle sampler. The NASA WB-57 carries a comprehensive aerosol and chemistry package; it again will sample the anvils and extend the measurements to 18 km. Its flights will also be optimised for validation of the Aura satellite. These two aircraft will concentrate on the edges of the anvils and the surrounding environment; the Egrett will provide the bulk of the measurements in the cirrus anvils. The ARA King Air will carry the UWA cloud lidar and a cloud radar, flying with the Egrett in the same manner as EMERALD-II; the aim is to penetrate thicker cirrus than last time using the radar where the lidar signal is unable to penetrate. Finally, the ARA Dimona (a low-level aircraft) will measure boundary layer turbulence and heat/moisture fluxes together with CO<sub>2</sub>. Clearly, the various aircraft will have to be closely coordinated for maximum scientific impact. Some of the TWP-ICE experiments will be designed for validating remote-sensing measurements (from satellite and ground) and will not be of direct interest to this proposal, while the Egrett latitudinal surveys (see below) will not be of interest to TWP-ICE. The bulk of the flying however will be concentrated on individual storms and cirrus anvils, and here the aircraft will be arranged to sample different altitudes and different parts of the storm outflow, as well as the background TTL. The combination of so many well-instrumented high-altitude aircraft will constitute a unique experiment and characterise the impact of convection on the chemical and microphysical state of the TTL to unprecedented accuracy.

In SCOUT-O3, the Geophysica and Falcon will fly together in the same way as the King Air and the Egrett (fig.1), i.e. with the Falcon lidar measuring vertical profiles along the Geophysica path. These aircraft will characterise the upper part of the TTL and the TLS (tropical lower stratosphere), with particular emphasis on water vapour and cirrus near the tropopause. The Falcon will also make a comprehensive set of chemical measurements at low level and in the convective part of the troposphere. If the SCOUT-O3 campaign is carried out in the pre-monsoon period, the measurements from this proposal would provide information on the water vapour, aerosol and chemical outflow from Hector while the Geophysica would be able to extend the vertical profiles into the lower stratosphere. Each experiment separately would provide unique new information about the TTL; together they will form a uniquely powerful combination.

## 4. Methodology and approach

### 4.1 Aircraft campaigns

The aircraft flights planned for this experiment fall into two categories:

- a) studies of the inflow and outflow from convection. First, the BAe146 aircraft will be flown in the boundary layer around the inflow to the chosen storm, sampling the aerosol and chemical fields in the inflow. Then, the Egrett will be flown into the outflow of the storm, guided by the King Air or the BAe146 lidar, as for EMERALD-II (TWP-ICE will provide the King Air). Different regions of the outflow will be studied in turn. During the first campaign (Nov-Dec 2005), we expect (on the basis of EMERALD-II) that the convection will be similar from one day to the next, so that the ensemble of flights can be used to build an overall picture of the anvils. During the monsoon campaign, by contrast, each storm will be different and the aim will be to sample a representative distribution of anvils by selecting storms with different characteristics (e.g. oceanic, land, isolated, cluster), to gain a statistical picture of anvil properties.
- b) Latitudinal surveys. The Egrett will be flown at maximum altitude (14.5 – 15 km, depending on conditions) on transects north and south of Darwin. Advantage will be taken of the transit flights from and to the Egrett's home base of Adelaide (35°S) for the southward survey. Northward transects will be necessarily more limited in extent, reaching about 5° north of Darwin, but will use different altitudes out and back. If available, the BAe146 lidar will be used to measure the vertical profiles of ozone, cloud and water vapour along the Egrett path. Otherwise, the BAe146 aircraft will be used to survey the low-level chemical and aerosol fields around Darwin when the Egrett is performing survey flights.

Both the BAe146 and Egrett aircraft will carry instruments for aerosol, chemical and tracer measurements. These are described in full in the Appendix. Most of the instruments we propose to use are available at one of the partner institutions, or as part of the aircraft facilities, or through collaboration. We propose to include four new instruments which will either be purchased, built or leased as part of this proposal:

- a) Cambridge will build two versions of the  $\mu$ -Dirac instrument which is a development of the DIRAC GC, flown successfully on the Egrett and on balloons (Robinson et al 2000). One will be deployed on the Egrett (50-80 samples / flight), with the other used on the ground to analyse samples from the whole air sampler on the BAe-146 (64 / flight). Here we will focus on CFC-11 (reduced concentrations indicate that stratospheric air is sampled), CH<sub>3</sub>CCl<sub>3</sub>, CCl<sub>4</sub>, CHCl<sub>3</sub>, Halon-1211 (all industrial tracers), CHBr<sub>3</sub>, CH<sub>2</sub>Br<sub>2</sub>,

and CH<sub>3</sub>I (an excellent tracer marine boundary layer air, Cohan et al 1999). Typical accuracy should be 5% or better, depending on compound. The anticipated sampling interval is 3-6 minutes.

- b) A Single Particle Soot Photometer (SP-2) to measure black carbon on the Egrett. This will be hired from DMT, USA. The SP-2 samples particles between 0.2 and 1  $\mu\text{m}$  in diameter and sizes them by optical scattering. The scattering signal is also used to gate a pulsed laser that heats the particle until it incandesces. The black body temperature is characteristic of the material incandescing. During ACTIVE we propose to fit the instrument into the U-Bay of the Egrett and sample using an inlet designed by Brechtel Manufacturing Inc. (BMI). The inlet uses a double diffuser cone and automatically varies the sample flow to maintain isokinetic sampling. Versions of this inlet are now in use on the NOAA P3 and the US DoE G-1 aircraft.
- c) GRIMM SMPS+CPC system (Model 5.402-900) will be used to measure submicron aerosol on the Egrett, also sampling from the BMI inlet. The instrument comprises an electrostatic classifier and CPC, capable of scanning between 15 nm and 900 nm mobility diameter. The instrument is robust and compact; however, the commercial instrument will not be capable of sampling at the reduced pressures that will be experienced. Modifications will be made, using the advice of Dr Hans Grimm, prior to the campaign.
- d) Forschungszentrum Julich NO/NO<sub>2</sub> instrument. This is a chemiluminescent instrument (reacting O<sub>3</sub> with NO) with an inherent sampling time of 10 Hz and a typical detection limit for a 4 s sample of 30-50 pptv. As its purpose is to measure NO<sub>x</sub> in the thunderstorm outflow this sensitivity is more than adequate. It is a version of the MOZAIC NO/NO<sub>y</sub> detector, supplied by Prof A. Volz-Thomas.

#### 4.2 Ozonesondes

During EMERALD-II, UWA ozonesonde equipment was taken to Darwin and used for a series of ozone profile measurements. These have been very useful in helping to interpret the aircraft ozone measurements. Given the overall focus of this campaign on the TTL region it is even more important that we fly ozonesondes to characterise the background atmosphere in which the flights are taking place. We intend flying one ozonesonde per flight day during the campaign periods (20 sondes) with extra flights when the background upper troposphere above Darwin in downwind of deep convection (10 sondes).

#### 4.3 Modelling and interpretative techniques

##### *Large scale modelling: TOMCAT*

p-TOMCAT is a new version of the three-dimensional global TOMCAT Chemical Transport Model (CTM). It has been parallelised using the MPI message-passing library and can be run on many tens of processors to achieve much improved performance. The model includes a detailed tropospheric chemistry scheme and includes photolysis, wet and dry deposition and a comprehensive set of emissions. The model also includes treatment of physical processes in the troposphere with parameterisations of shallow and deep convective transport, boundary layer mixing and vertical diffusion. The model has been verified by several comparisons against observations from established datasets and during measurements campaigns. Recently, the improvements to the performance of the model have enabled some preliminary integrations at very high resolutions (0.5°x0.5°) to be performed. The model will be used, along with trajectory studies (see next), to diagnose transport into and out of the TTL by a variety of processes. p-TOMCAT will provide information on the expected variability of the TTL and possible differences between the two proposed campaign periods. We expect this new dataset to point to important deficiencies in model treatment of this region.

##### *Trajectory modelling*

The use of trajectory calculations based on 3-D winds from global meteorological datasets is well established as an approach to interpreting and complementing in-situ chemical data. Recent studies in the extratropical troposphere indicate very good consistency between calculated trajectories and structure observed in chemical fields, supporting the idea that the wind fields are sufficiently accurate for this purpose (e.g. Methven et al 2003). Dynamical phenomena in the tropics are probably not so well resolved by the meteorological datasets and such trajectories cannot capture all aspects of non-convective transport. However agreement between structure measured in water vapour, for example, and calculated trajectories is often very good (J. Methven, personal communication), suggesting that when used with due care, taking account of the known presence of convective systems, they can be a valuable complement to observations. The Cambridge group have significant experience in use of trajectory calculations to study the tropical tropopause region (Bonazzola and Haynes 2004) and also in chemical calculations along trajectories (Evans et al 2000).

##### *Large Eddy Modelling*

A combination of the Met Office Cloud Resolving Model (CRM) and the UMIST explicit microphysics model (EMM) will be used to simulate individual storms. This approach is currently being used to interpret the

EMERALD-II dataset and was successfully used by Phillips et al (2001) to model storms forming over New Mexico. This model will provide the main theoretical tool to link the aerosol and microphysical measurements by the two aircraft.

The CRM includes parameterised microphysics with a double-moment scheme representing the number density and mixing ratios of the ice phase as described by Swann (1998). UMIST will run this model for each of the observational case studies using aircraft and radiosonde ascent data as input. The cloud structure predicted by the model as the storm develops will be compared to the radar image, i.e. the model will predict the radar reflectivity. Sensitivity tests will be performed with the model, varying input parameters over the range of their uncertainty e.g. wind shear, temperature profile and ice nucleus concentration. This will enable us to understand the sensitivity of the model to each of these parameters and hence determine the reason for differences between the predictions and observations.

The 3-D cloud dynamics and cloud water content from the CRM simulations provide the input parameters for the EMM. This model treats the following microphysics explicitly within this dynamical framework:

- a. The activation of cloud droplets from the aerosol entering the cloud.
- b. The growth of raindrops by collision coalescence, the production of ice particles by primary nucleation.
- c. The homogeneous nucleation of ice is treated at temperatures below  $-35^{\circ}\text{C}$ .
- d. The growth of ice particles, of habit determined by temperature and supersaturation (including ventilation effects) aggregation and riming.
- e. Secondary ice particle production by riming splintering, raindrop freezing and ice crystal evaporation.
- f. The melting of precipitation below the freezing level is treated along with the recirculation of hydrometeors between neighbouring updraughts and downdraughts.

In this way the model is able to predict the microphysical properties of the anvil region for direct comparison with the in situ measurements. It is also able to predict the fluxes of water vapour, particles and passive tracers out of the cloud through the anvil region. The detailed microphysical model will also enable direct comparisons between the modelled evolution of the cloud and measurements of reflectivity from the radar both as the cloud develops and in its mature state.

#### *Microphysics, Aerosol and Chemistry (MAC) model*

The specific advantage of MAC in this project is that it simulates a size-resolved aerosol interactively with the cloud microphysics and dynamics, which is not done in the EMM. MAC currently uses a 2-D axisymmetric/slab-symmetric configuration for the dynamics with bin-resolved microphysics for aerosol, drops, ice, graupel and aggregates (Yin et al 2001, 2002, 2004). It also includes a kinetic treatment of gas scavenging and bin-resolved aqueous-phase chemistry. Therefore, size-resolved aerosol particles processed by clouds and detrained from the cloud outflow region can be simulated. Extension of the model to a 3-D version and to include cloud electricity is underway and will be complete before the field campaign. This model will be the main theoretical tool for investigating chemical transport and generation ( $\text{NO}_x/\text{O}_3$ ) in the storms. It will also be used to interpret the black carbon measurements. For the lightning scheme, measurements from the Osaka University lightning interferometer at Darwin will be used to constrain the simulations.

Simulations with MAC can include the effects of variable aerosol composition and externally mixed aerosol distributions, which can influence cloud droplet spectra. On the other hand, the formation and diffusional growth of ice particles, and the production of ice by secondary processes such as the Hallett-Mossop process, are calculated in more detail in the EMM. Simulation of the same case studies using the two models, with regular intercomparisons, will enable us to reach more robust conclusions regarding the microphysical and dynamical response to aerosols.

In the second (monsoon) campaign we expect to encounter a much greater variety of convective clouds than in the pre-Christmas campaign. It is not feasible, nor useful, to simulate them all - it is the ensemble properties of these clouds that will be of interest. The BAe146 measurements will provide the range of aerosol concentrations and types entering the storms (bearing in mind that Darwin, being a coastal location, is within reach of convection over land and ocean). Thermodynamic profiles will be available from the network of radiosondes as well as the BAe146 and other TWP-ICE aircraft. These will be used as input to the model to examine the range of anvil properties (aerosol and cirrus and chemicals) that it produces; these will be compared with the observations and used to improve the model simulations.