APPRAISE Final Report

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1. Basic Overview of APPRAISE

The Aerosol Properties, Processes And InfluenceS on the Earth’s Climate (APPRAISE) programme was a UK NERC (Natural Environment Research Council) directed research programme designed to improve our ability to quantify the effects of atmospheric aerosol particles on the Earth’s climate system. The aims of APPRAISE were to investigate and understand the underlying properties of airborne particles that affect the lifecycle of aerosol particles which in the end are responsible for their effect on radiation and cloud formation (hence the aerosols direct and indirect radiative contributions to the earth’s radiation budget). The programme tackled the formation, transformation and interaction of particles in the atmosphere to establish and quantify key pathways in their lifecycle.

The full scientific objectives of APPRAISE are described in detail in the APPRAISE Science Plan, which can be obtained from the NERC APPRAISE website at: http://www.nerc.ac.uk/research/programmes/appraise/resources.asp or from the APPRAISE website at NCAS (the National Centre for Atmospheric Science – A NERC research centre) http://ncasweb.leeds.ac.uk/appraise. The Science Plan also outlines the areas of research that APPRAISE intended to fund. The APPRAISE Implementation Plan (also available from the above web sites) sets out the structure of the programme, its management, its knowledge exchange and its links with other research programmes within NERC, and in the UK and international communities. The Implementation Plan thus sets out the strategy the programme intended to follow in order to achieve its scientific objectives as laid out in the Science Plan. The information contained in each of these documents will therefore only be briefly summarised here.

1.1 The key objectives of APPRAISE

Over its lifetime, APPRAISE sought to tackle key research challenges and aimed to better understand and quantify:

1. the direct effect of aerosols on the Earth’s radiation budget, via scattering and/or absorption of radiation;
2. the influence of aerosols on cloud properties and hence indirect effect on climate and influence on the hydrological cycle;
3. the role of aerosols in feedback processes between land, the biosphere and climate.

It was realised that to reduce uncertainties in these areas required a much-improved understanding of the atmospheric aerosol lifecycle including better representations of this in climate simulations.

With this in mind, the main aims of APPRAISE were to:

Provide a focus for UK research in the key area of aerosols, clouds and climate over a 5 year period

- Deliver improved knowledge on aerosol and cloud properties, processes and impacts through detailed field and laboratory measurements
- Use process models to develop parameterisations for use by global and regional transport and climate models.
- Test and demonstrate the climatic importance of key aerosol and cloud processes through a coordinated hierarchy of models linking process scales to global climate models through transport models
- Identify and quantify key biosphere-atmospheric aerosol couplings at the process scale and so provide improved data from which to develop parameterisations.
- Deliver a coordinated internationally leading activity in the APPRAISE area
In achieving these aims APPRAISE contributed to meeting the challenges firstly within NERC’s Delivery Plan 2005-2008, and then to those within the newly set up (soon after the start of APPRAISE) NERC science themes. For example, by using targeted process studies, the observational data set was improved and expanded to enable use to validate climate models, which was a key challenge of the Climate System theme.

1.2 The APPRAISE strategy

To significantly develop knowledge of the impact of aerosols on our climate APPRAISE undertook scientific research to examine the key processes involved in the lifecycle of aerosols in the atmosphere. This was carried out through the construction of an integrated hierarchy of laboratory studies, field and global observations, process models, regional models and global/climate models, to provide interconnected tools for use in three key science areas. The mechanism for delivering this research was through the funding of consortium grants in each of these three key areas, supported by a core programme of key strategic scientific activities.

Consortium projects were funded to target investigation of:

- The Direct Impacts of Aerosol on Climate
- Aerosol Cloud Interactions and Climate
- Aerosol coupling in the Earth System

The successful proposals in these three targeted areas of research respectively were:

a) APPRAISE-ADIENT  (PI: Prof. Don Grainger, Oxford University)
b) APPRAISE-Clouds  (PI: Prof. Tom Choularton, University of Manchester)
c) APPRAISE-ACES  (PI: Prof. Gordon McFiggans, University of Manchester)

However, to achieve its overall objectives, add value to the consortia activities and provide a strategic vision for aerosols research, APPRAISE also supported several key scientific areas of research that it saw as providing underpinning science to the programme. These specifically targeted research areas were not covered by funding and activities within the NERC institutes and facilities (e.g. NCAS), although close collaboration with these entities was achieved throughout the lifetime of the programme. The targeted core research areas funded within APPRAISE were:

1) Formation Pathways and Properties of Organic Aerosols
2) Improved Representation and Validation of the Radiative Properties and Impacts of Aerosols and Clouds
3) Development of a Global Model to Investigate Key Aerosol and Cloud Processes
4) Improved Coupling of In Situ Cloud Measurements with Novel Model Schemes

Applications to undertake this programme of core research saw successful bids in each of the areas 2-4, while 2 successful bids provided complementary research options in area 1.

Highlights of the main scientific achievements within each of these core areas and from the consortium projects will be given in section 3 of this report. However, the more generic achievements of Appraise will be discussed first in section 2.

The format adopted within APPRAISE, of initially funding a core programme of key research work which could then feed into the targeted research areas undertaken within the funded consortia projects, proved to be highly successful. Each of the core projects developed a suite of (modelling or analysis) tools which were initially developed using and/or tested on existing data sets or samples, but which were then used extensively throughout the consortia research work that followed. Apart from being a significant research output in their own right, in all cases the core research activities continued within the consortium projects and form an important legacy of the programme.
As envisaged, the work undertaken within the core projects significantly underpinned the targeted research which followed in each of the consortium projects. For example, the tools developed to trap individual aerosol particles by optical tweezer and electrodynamic balance techniques in CP1a (see section 3.1), were used to measure the uptake or release of water vapour and organic species to/from (and in some cases the reactivity of) aerosols of various composition. This information on the hygroscopicity, volatility and reactivity of these aerosols was then used extensively within the ACES consortium project, to update rates etc in models used to understand and interpret measurements made both in the rainforest in Borneo and in the chamber studies which followed in Manchester. Similarly, the techniques developed in CP1b to investigate the composition of low molecular weight and heterogeneous reaction product organic aerosols, were used extensively to analyse the ACES samples, as well as samples collected during the APPRAISE-Clouds consortium project. These results fed back into the development of the Master Chemical Mechanism in ACES. CP2 developed a hierarchy of radiation transfer models and an aerosols optical property database which were used to process the aerosol data sets collected in particular during the ADIENT project (as well as other pre/post APPRAISE data sets). The family of models of differing complexity generated in CP3 to improve the representation of aerosols in Global Climate Models also proved useful in developing the results obtained in the various consortia field experiments. The detailed model of aerosols and clouds developed in CP4 and adapted to operate in various dynamical frameworks, was used extensively to understand and interpret the measurements obtained during APPRAISE-Clouds in the array of cloud types measured, as well as the AIDA and MICC cloud chambers. In most cases, the use of core project analysis tools, techniques and models within the consortium projects also led to steady development and improvement of these techniques throughout the APPRAISE programme.

2. The Generic Achievements of APPRAISE

The APPRAISE directed research programme was designed to fund targeted and strategic activities in the field of aerosol science, and provide a platform for focussing and coordinating NERC’s existing and future investments in this area. The aim was to maximise the potential of the NERC community’s existing expertise and build links between science activities that were already internationally competitive. Hence APPRAISE and its investigators would continue to collaborate with the Hadley Centre, and would complement the aerosol research about to be underway or underway in parallel research programmes such as UK-SOLAS (NERC’s UK Surface-Ocean, Lower-Atmosphere Study) and QUEST (Quantifying Earth system processes and feedbacks for better informed assessments of alternative futures of the global environment). APPRAISE would continue existing collaborations with the Centre for Ecology and Hydrology (CEH) and NCAS (then the NERC Centres for Atmospheric Science - now the National Centre for Atmospheric Science) programmes, and fully utilise experimental facilities such as the (then new) state-of-the-art research aircraft facility (FAAM), instrumentation and synergistic links to existing modelling programmes. It was intended that this would allow APPRAISE to develop the aerosol area of NERC’s science strategy into an internationally leading activity, providing a framework, within which UK science could work collaboratively with International programmes and scientists.

In summary, APPRAISE would provide key information on aerosol and cloud properties, processes and effects on climate, which could then be used to further develop Global Climate models, particularly the Hadley Centre’s climate model, enabling improved representation of aerosol and cloud processes in such models and thus helping to lead to a reduction in uncertainty in climate change predictions and feedbacks. By working closely with the Hadley Centre, the components of NCAS (initially these were the Centre for Global Atmospheric Modelling (CGAM) and the Distributed Institute for Atmospheric Composition (DIAC)), APPRAISE would ensure effective transfer of information from the programme into the global climate models.
At the end of the APPRAISE programme it is evident that the majority of these aspirations and goals have been achieved, and in some areas surpassed. One of the main achievements of the APPRAISE programme has been to assemble a group of world leading investigators and modellers of aerosols and their properties (some of whom prior to APPRAISE were not even aware of each others work) into a fully collaborative grouping, fully accustomed to working with each other, and capable of going on to tackle the remaining big questions in the aerosols and clouds field. Prior to APPRAISE, while a lot of good research was being undertaken by individuals or smaller groups, these were often not capable of looking at the “Bigger picture” of aerosol properties, processes and influences on the earth’s climate in a fully integrative way. This is now not the case, and a new community of aerosol scientists, including a group of newly trained and highly motivated young investigators, has been built up. Within this group it is now almost second nature to collaborate with one another to overcome the challenges within aerosol science as they become priorities. This is a significant achievement of APPRAISE. A measure of this outcome can be seen by looking at the investigators and institutes leading research within some of NERCs new research programmes, which exist to tackle the questions identified as being important within NERCs seven science themes. As an example, the majority of lead investigators within NERCs new Aerosols and Clouds programme are comprised of people who played a leading role in the APPRAISE core science work and/or the APPRAISE consortia projects, or indeed involved in APPRAISE KE projects/activities. In fact, the new Aerosol and Clouds programme aims to build on much of the scientific progress achieved within APPRAISE, concentrating on a programme of laboratory investigations, modelling and model development. A similar picture is also seen within other programmes like the Arctic Research Programme, or the Storms Risk Management programme, which are less obviously an “organic” development from APPRAISE. In these programmes, the collaborative habit attained within APPRAISE is strengthened by a subset of APPRAISE investigators who are forging new links with investigators in completely new areas of research, and well outside the original scope of APPRAISE.

In addition to building a UK aerosol community, the APPRAISE programme also succeeded in developing existing and new links through collaboration with a host of external institutes and programmes undertaking similar or complementary work. As envisaged at the outset, close collaboration with the Met Office continued during APPRAISE, particularly through the work carried out to include aerosols and their properties in Global climate models. Unsurprisingly, this collaboration was most notable through work undertaken within core project CP3, which built a model framework to investigate what key aerosol and cloud processes, and in what degree of complexity, were important to be included in regional and global scale models. Through the development of both bin and modal representations of aerosols, work progressed sufficiently within their GLOMAP model framework to enable the GLOMAP-mode module to be included in the UK Cloud and Aerosol (UKCA) model now embedded in the Met Office HADgem3 climate model (see CP3 highlights). In CP2, a hierarchy of radiation transfer models was developed for testing and comparison with Global Model Radiation schemes. Model development within CP4 and the APPRAISE-Clouds consortium has led to a new understanding of ice nucleation on dust particles, and of the glaciation of clouds in different circumstances. A considerable programme of field measurements using the FAAM BAe146 aircraft was also undertaken in APPRAISE-Clouds in close collaboration with the Met Office OBR group who were interested in investigating similar cloud processes.

Other significant achievements of the APPRAISE programme include the undertaking of significant field and laboratory studies and experiments. This occurred within each of the 3 consortium projects. The ACES consortium participated in and made a significant contribution to the NERC OP3 (Oxidant and Particle Photochemical Processes above a South-East Asian Tropical Rainforest) consortium project in Borneo during April-July 2008. Measurements of gaseous and aerosol concentrations and fluxes, were made above and below the rain forest canopy and in an oil palm plantation. These field measurements were
used to guide a programme of accompanying laboratory investigations of secondary organic aerosol (SOA) in the Manchester photochemical chamber, and to further develop model tools. Research highlights from these investigations are presented in section 3 of this report.

Both ADIENT and APPRAISE-Clouds consortia made significant use of the FAAM research aircraft to undertake a whole series of flights within and around the UK and over NW Europe. ADIENT made airborne measurements of aerosols and trace gases around the UK and downwind of urban conurbations characterising aerosols and their properties close to source and as a result of aging. These were undertaken in close collaboration with the Met Office OBR radiation group. Through links with the EU sixth framework European Integrated project on Aerosol, Cloud, Climate and Air Quality Interactions (EUCAARI), ADIENT participated in the EUCAARI-LONGREX experiment in May 2008 (which involved multiple aircraft making remote sensing and insitu measurements) undertaking long range studies of the concentrations, composition and aging of aerosols across the whole of NW Europe. As well as an extensive data set from the UK 146 aircraft (on BADC) this provided links to the full EUCAARI data set (held at NILU) including the EUCAARI-IMPACT experiment data set (concerned mainly with cloud-aerosol-radiation interactions near the Cabauw site in Netherlands) as well as an EMISSIONS data base. Scientific highlights are provided in section 3 below.

APPRAISE-Clouds undertook 28 science flights using the FAAM BAe146 aircraft, 4 of which were over Switzerland and Germany during EUCAARI in May 2008. 21 flights were conducted over the SW of England in the vicinity of the remote sensing radars and lidars at the STFC Chilbolton Facility for Atmospheric and Radio Research (CFARR), where the consortium also deployed the FGAM instrumented sea-container laboratory to conduct a number of detailed ground based aerosol measurements during the APPRAISE–Clouds intensive observation period (IOP) from December 2008 to March 2009. Many of the 146 flights were undertaken in close collaboration with the Met Office OBR cloud physics group. CFARR also undertook a long term programme of observations of the development of the ice phase in clouds of varying type passing overhead as part of the APPRAISE-Clouds project. In addition, through links with the Karlsruhe Institute of Technology (KIT), APPRAISE-Clouds participated in a programme of laboratory studies of aerosols and their ice-nucleating properties at the AIDA Ice chamber facility (which is a EUROCHAMP-2 facility), and continued complementary investigations at the Manchester Ice Cloud Chamber (MICC). Highlights from these experiments are also summarised in Section 3.

The data sets derived from the field and laboratory work carried out within each of the 3 APPRAISE consortia are held at the BADC (with links to collaborative data bases and data sets). These form part of the legacy of the APPRAISE programme (see section 5), along with the framework of models developed to help to understand aerosol properties, processes and evolution, to evaluate their radiative contributions and to prioritise these for inclusion in larger regional and global scale climate models.

In discussing the achievements of the APPRAISE programme, the organisation of annual science meetings should not be overlooked. These enabled the achievements in each sphere of the APPRAISE programme to be communicated to the rest of the community. By inviting international keynote speakers who were world renowned experts in the different arenas of aerosol and cloud science, it was also possible to promote the APPRAISE achievements to the outside world, and to see how these achievements fitted into the international view of aerosol and cloud science. Of course, the publication of peer reviewed papers and the presentation of results at key international conferences and workshops played its usual significant role in disseminating the main achievements of the APPRAISE program to the outside world. Alongside the data sets and modelling tools, the publication list of peer reviewed papers (see Appendix 1) is another of the main legacies of the APPRAISE programme. An updated will be available on the NCAS APPRAISE website.
3. The main scientific highlights of the APPRAISE programme.

3.1 Highlights from the APPRAISE Core Projects:
**CP1a: Formation Pathways and Properties of Organic Aerosols** (PI Dr Jonathan Reid, University of Bristol)

- **Measurements of Condensation and Evaporation of Water on Single Aerosol Particles.**
  
  One of the largest uncertainties contributing to our lack of confidence in quantifying the indirect influence of aerosols on climate is the rate at which water can condense on or evaporate from atmospheric aerosol. The condensation of water on aerosol is the key process that leads to the formation of cloud droplets and uncertainty in the rate has a large impact on the number concentration of cloud droplets that can form.

  In Bristol through support from APPRAISE, a novel method for measuring the rate of water condensation or evaporation from aerosol was developed (using an optical-tweezers method to trap the particles) to help resolve this issue, quantifying droplet size changes of less than 1 nm to measure directly the deposition of water at a particle surface. In particular, measurements are now being made to quantify the influence of organic components on the condensation rate.

  ![Figure 1](image1.png)

  **Figure 1:** The time dependence of particle size can be followed with sub-nanometre accuracy (see insert) during the condensation process as the aerosol approaches an equilibrium state.

- An important question is - How accurately can critical supersaturation ($S_c$) be measured?
- At high RH (Fig 2, right): Need to understand errors associated with equilibrium properties (solute effect) on coarse particles to measure and predict $S_c$ for accumulation mode.
- NaCl/Ammonium Sulphate is used to calibrate instruments for determining $S_c$ etc.

- Condensation/evaporation limited by surface kinetics
- Timescales for equilibration can become extremely long as the glass transition is approached - depends on magnitude and direction of RH change (Fig 3 right).
- At high organic concentration, organic rich aerosol can undergo a glass transition, becoming highly viscous.

  e.g. Diffusion profiles of water into a highly viscous sucrose particle lead to steep diffusion “fronts”
Complementary single particle studies undertaken at Cambridge University (co-I Francis Pope) using an Electro Dynamic Balance technique (Fig 4, right) looking at: (i) Hygroscopicity and volatility of (particularly organic) aerosols; (ii) Reactivity of aerosols; (iii) Primary biological particles e.g. pollen

- Humic acid: organic with very high hygroscopicity per molecule (but the molecules are very large)
- Volatility of dicarboxylic acid liquid phase has been measured; but needs reconciliation between liquid and solid phase studies.

- Aerosol reactivity can be phase dependent.
- Aerosol composition affected by phase dependant oxidation.
- Pollen spores (Fig 4, middle right) can potentially act as giant cloud condensation nuclei (e.g. external uptake of water: Fig 4, bottom right).

CP1b: Investigating the Composition of Organic Aerosols: Low Molecular Weight and Heterogeneous Reaction Products. (PI-Dr Alastair Lewis, University of York)

A key to understanding properties of aerosols and their evolution is a determination of their chemical composition, particularly that of the key organic species (out of the myriad of potential organic components) that could contribute.

At York, core project CP1b applied, and further developed, novel state-of-the-art analytical molecular characterisation methods, first by analysing existing available aerosol and smog chamber experiment data sets/samples, (in preparation for the analysis of consortium project samples as they were to come on-line). This determination is difficult because of:

- Complexity - Emission sources range of molecular weights, polarities, high number of hetero-molecules (i.e. Cl, Br, N, O, S)
- Primary hydrocarbon emissions - As molecular weight increases, the number of possible isomers increases exponentially.
- Secondary Organic Aerosol (SOA)
  - Highly oxidised and composition largely unknown
  - Huge numbers of compounds at very low concentrations
  - For many cases – no authentic standards for comparison

Determination requires use of a range of complementary techniques:

1. Determination of key low molecular weight monomer building blocks:
   - Volatile and semi-volatile polar compounds in SOA analysed using comprehensive two dimensional gas chromatography and mass spectrometry (GCXGC-TOF/MS). High resolution.

   - Separates by volatility and polarity (Fig 5a, below left).
- Release organics from OA by thermal desorption
- Converts data to a 3D contour plot or surface diagram. Each spot represents an individual compound (Fig 5b, below right)

2. High molecular weight analysis:
- Resolve the molecular structures e.g. of oligomeric compounds in SOA formed via heterogeneous reactions of monomers (measured as above)
- using high performance liquid chromatography coupled with tandem mass spectrometry (e.g. MS-MS of Cyclohexene SOA monomers and O3 in dark in EUPHORE chamber - Fig 6(a, b), below left & right)

Development of these extremely useful techniques has allowed:
- determination of aerosol composition;
- identification of new reaction pathways identified for comparison with model (MCM – Master Chemical Mechanism) output
- determination of links to aerosol properties (e.g. hygroscopicity, volatility)
- potential for source identification.

Through interaction with the APPRAISE consortia and external collaborations, the goal was to link SOA compositional data obtained in simulations with that found in the real atmosphere in a range of locations, ranging from urban to the tropical rainforest, e.g. using LC-MS$^n$ studied the SOA from a-pinene ozonolysis and compared the SOA composition and formation modelled using the Master Chemical Mechanism (Univ. Birmingham and Leeds)
CP2: Improved representation and validation of the radiative properties and impacts of aerosols and clouds. (PI - Dr Eleanor Highwood, University of Reading)

The direct effect and radiative forcing of atmospheric aerosols depend on properties such as extinction co-efficient, single scattering albedo and asymmetry parameter. These in turn depend on aerosol properties such as size, composition and hygroscopicity etc. A key requirement of the APPRAISE programme was to gain a better representation of these measured properties of aerosols in models which could then be used to calculate the optical properties of aerosols in large scale climate models.

In CP2, tools and frameworks were developed at Reading to improve the representation of aerosols in radiative and climate models. This produced:

- Development of a hierarchy of radiative transfer models, including a benchmark radiative transfer model (RFM DISTORT).
- Testing and favourable comparison with GCM radiation schemes for a number of real and idealised cases (including comparison with models in the international AEROCOM framework)
- Validation against lab and satellite data
- Comprehensive documentation for methodology, models and test case results
- An aerosols optical properties database (from models and observations) [see http://www.met.reading.ac.uk/adient/refractiveindices.html] developed alongside UKAAN

- Provision of processing software/documentation for producing APPRAISE aerosol products from FAAM data at BADC
- Potential for processing pre-existing FAAM datasets (on request)
- Continued development to expand scattering framework to include codes for non-spherical scattering (e.g. post APPRAISE development within FENNEC)
CP3: The Development of a Global Model to Investigate Key Aerosol and Cloud Processes (PI - Professor Ken Carslaw, University of Leeds)

This core activity at Leeds University had 2 main objectives:

1. To provide a global aerosol transport model capable of:
   • carrying the main aerosol types,
   • dealing with complex mixing states
   • acting as a key stage in the hierarchical testing of aerosol processes in APPRAISE

2. The coordination of the hierarchical model development, interfacing this with other modelling efforts in the UK, e.g. comparing & ultimately improving UKCA in the Unified Model
   • The aim was thus to develop models capable of carrying the essential properties of aerosols and their interaction and evolution within cloud so that these effects could be properly investigated on the global scale whilst still remaining computationally affordable.

To this end a family of models in which the aerosols were represented with differing degrees of complexity were compared with each other in different model environments; e.g. two representations of the aerosol size distribution within GLOMAP (GLOBal Model of Aerosol Processes) were run within a fairly complex 3D offline Chemical Transport Model (CTM) and compared. A more complex sectional (or bin) representation (below left) and a simpler modal representation (below right) of the aerosol size distribution were produced and run within the CTM framework and then compared under different scenarios.

Fig 8: GLOMAP-bin aerosol number & mass in size bins (sections); GLOMAP-mode: aerosol number & mass in log-normal modes. Fig 9 (below) shows the different model complexities.

The UKCA (UK chemistry and aerosol global model) uses the GLOMAP-mode aerosol scheme, coupled to tropospheric and stratospheric chemical schemes within the Met Office HadGEM3 climate model. The CTM was used as a test bed for improvements to the climate model aerosol scheme and to investigate the role of specific processes. The aerosol schemes were developed to be capable of dealing with the major aerosol types and complex aerosol mixing states.

The UKCA aerosol scheme carries approximately 20 tracers whereas GLOMAP carries more than 100. The microphysical processes are essentially similar. Hence, UKCA is ideal for multi-year simulations, whereas GLOMAP is ideal for detailed process studies (where size distribution is key) of the order of months.
After development in CP3, these models were used to evaluate understanding of large scale aerosol microphysical, chemical and radiative properties against observations, and for developing and testing parameterized schemes that captured the key processes, but which were simple enough to be incorporated into global and regional climate models.

Key model developments for GLOMAP-TOMCAT included:

- improved treatment of secondary organic aerosol
- calculation of aerosol thermodynamics for inorganic/organic mixtures
- treatment of additional particle types
- multi-stage grid nesting down to 10x10 km.

Standard model diagnostics were developed to allow comparison with a wide range of aerosol instrumentation.

The core project concluded (in September 2009) having completed the planned development activities and moved into a phase of model exploitation. Achievements included:

- New code to calculate size-resolved uptake of nitrate and ammonium using a hybrid kinetic/equilibrium approach developed and tested; produced very nice results.
- Calculation of the effect of wind speed changes on marine aerosol and climate:
- New understanding of the impact of nucleation on global aerosol and climate:
- Testing a new nucleation mechanism in a global model, highlighting the importance of organics in nucleation and particle formation.
- New chemistry module to quantify the effect of Br chemistry on DMS and aerosol
- First results with a dust module compared with observations, showing that dust has little effect on ambient CCN.
- First test of a cloud drop number model.
- A new chemical composition code implemented (see above)
- New code to handle aerosol scavenging in drizzle developed and tested (e.g. for Arctic conditions where model performance was previously very poor).
- An extensive database of CN measurements has been compiled and the model evaluated
- An extensive database of CCN concentrations has been compiled and the model evaluated

Work with the Met Office continues. The GLOMAP-mode model implemented in UKCA is still being updated based on findings from the offline model (GLOMAP-TOMCAT). E.g. ultrafine sea-spray and boundary layer nucleation being included based on research in other NERC projects.

(PI - Professor Tom Choularton, University of Manchester)

Work in this core area was performed jointly by the School of Earth Atmospheric and Environmental Sciences at the University of Manchester and at the School of Earth and Environment at the University of Leeds. The resulting APPRAISE Core model framework consists of a range of increasingly detailed and complex descriptions of the aerosol and cloud processes set within a dynamical framework of varying complexity (and hence computational cost) to enable calculation of the aerosol and cloud processes at the required level of detail.

The most detailed bin size distribution representation of the aerosol and cloud particles exists within a one dimensional column model framework, although another version of the model has been developed to run in a Large Eddy dynamical framework. The latter can also incorporate a bin microphysical representation, but some of the detail of the treatment of the aerosol chemistry and microphysics is reduced. Hence this detailed core APPRAISE model (known as ACPIM) exists essentially in two main forms for application in different situations e.g. for use in chamber and field studies.

The detailed model:

- solves for the growth of aerosols (with multi-component chemical species); cloud drops; and ice particles (of varying density).
- has only a single class of particles, the aerosols; all other properties are characteristics of the aerosols; e.g. the water content determines whether the particles are cloud drops or rain particles; their phase of matter along with the particles growth trajectory determines whether they are liquid or ice.
- has the capability to simulate the effects of aerosols on cloud properties; the formation of rain; snow; sleet; and hail.

A main requirement in CP4 was to develop a model scheme capable of taking input from advanced aerosol models, such as the Aerosol Diameter Dependent Equilibrium Model (ADDEM) developed previously in Manchester. It is restrictive to use the full ADDEM code within the APPRAISE core model, since it is computationally very expensive; so the approach has been to use polynomial fits to the aerosol thermodynamic equilibrium properties for each chemical species. For internal mixtures, the Zdanovski-Stokes and Robinson (ZSR) approximation was assessed to be sufficient since it gives very similar results for simple systems. The full model incorporates:

- A description of the heterogeneous freezing and the heterogeneous deposition processes (developed through working closely with Dr Ottmar Moehler of the Forschungszentrum Karlsruhe in Germany, who works in the AIDA cloud chamber facility). A new parameterisation for freezing was also developed (published in ACP in 2009).
- ice crystal growth by vapour diffusion and riming along with droplet growth by vapour diffusion and coalescence.
- a new treatment of ice crystal aggregation based on experimental studies performed in the Manchester Cloud Chamber and modelling studies at the University of Reading.

Another bin-resolved microphysics model set within a large eddy model (LEM) framework has been developed at The University of Leeds. This new model consists of four hydrometeor species, namely, drops, ice crystals, graupel and snow. The model incorporates
a bin scheme based on the Microphysics, Aerosol and Chemistry Convective Cloud Model (MAC3) developed in Leeds.

Both versions of the APPRAISE cloud model have been successfully applied to understanding the development and evolution of the clouds investigated in a number of lab studies and field projects, including the APPRAISE-Clouds consortium project. As an example, the detailed column model was used to investigate the formation of droplets and the initiation of ice in convective clouds. These cloud types are particularly suited to this kind of analysis. In this case, the aerosol size resolved composition measured in detail at the surface was input to the model (after confirmation that it was representative of the air entering the convective cloud through its base by comparing with the more limited suite of aerosol measurements conducted on the aircraft flying just below cloud). The effect of simulating the cloud development and its sensitivities to various aerosol properties proved to be very a powerful tool to understanding the formation and development of ice and precipitation in these and other cloud types.

The LEM version of the model has been used as a tool to enhance understanding in other cloud cases. Supercooled stratocumulus clouds observed during one APPRAISE-Clouds (18 February 2009) case contained small amounts of ice. The new model was applied and good agreement seen between the simulated and observed clouds. The model reproduced two cloud layers, an ice cloud layer between 2 and 4 km, and a water cloud near to the ground. The simulated cloud top at about 4 km was close to the observed top and the simulated concentrations of ice particles were similar to the observed values even though the concentrations were high for such high temperatures. Similarly, the LEM version of the model has been used to elucidate the detail of the dynamics and microphysics of mountain wave clouds investigated in other cases.
3.2 Highlights from the APPRAISE Consortium projects:

- **Aerosol Coupling in the Earth System (The ACES consortium)**
  (consortium led by Prof. Gordon McFiggans, University of Manchester with colleagues from the Universities of Manchester, Edinburgh, Leicester, Lancaster, York and Imperial College London, and the Centre for Ecology and Hydrology).

**Summary**

The Aerosol Coupling in the Earth System (ACES) consortium has led to major strides forward in understanding the cycling of biogenic aerosol and its precursors in the tropical environment. Synergistic collocated deployment of targeted instrumentation within the OP3 project has enabled a comprehensive *in situ* characterisation of ambient aerosol in the Bornean rainforest. The field measurements, along with a vegetation survey, were used to guide a programme of photochemical chamber secondary organic aerosol (SOA) investigations using individual or simple synthetic precursor compounds and real emissions from mesocosm experiments. The field and chamber experiments provided both input and measurement constraint for the development of oxidative degradation mechanisms for selected biogenic volatile organic compounds (VOCs) that have, in turn, been used to simulate the chamber SOA formation and transformation. Large-scale models, constrained by satellite observations have been used to construct aerosol budgets over S. Asia for comparison with *in situ* observations.

**Field results**

A major summer field campaign was conducted in conjunction with the OP3 consortium in a tropical rain forest in the Danum Valley, Sabah-Malaysia, Borneo in 2008. A unique and broad data set of gaseous and aerosol concentrations and fluxes, both above and below the forest canopy and in an oil palm plantation was collected. The ACES data largely, but not solely, focussed on in-canopy and oil palm aerosol and VOC measurements. Some of the highlights are outlined below.

Leaf level screening of 31 vegetation species in the GAW tower footprint revealed that 16 of species contribute 78% of vegetation, allowing focus to be placed on dominant VOC emitting species. A biomass weighted average terpene emission factor was deduced from the screened species. Leaf level screening at the oil palm plantation yielded isoprene and terpene emission rates. In-canopy measurements of VOCs indicate that at least 90% of the monoterpene emissions are light-dependent, in contrast to those from temperate vegetation. Continuous profiles of aerosol size distribution, temperature and humidity were made along with cloud-fog water content in-canopy and above the forest.

![Profile measurement set up (left) and some particle concentration data (right), showing Fog formation in the upper canopy at night, and biological aerosol production in the under-storey during daytime](image)

A novel single particle detection system was deployed to attribute the aerosol number budget between total and viable biological material. Aerosol samples were collected as a function of
height in-canopy and analysed for size dependent composition by aerosol mass spectrometer (AMS). In-canopy mass spectral fingerprints were found to be broadly the same as those measured above and on the FAAM aircraft at the same location. Using samples from the Particle into Liquid Sampler (PiLS) at the rainforest site, with a method developed to analyse water soluble organic carbon, identifying 5 tracer compounds for SOA, biomass burning (BB) and direct plant emissions.

Oil Palm isoprene emissions were quantified and found to exceed rain forest emissions by a factor of 4, while monoterpenes emissions are negligible. For the first time, circadian control of isoprene emissions was derived (using canopy-scale flux measurements). Furthermore, oil Palm was found to be a significant emitter of estragole, a BVOC which has only recently started to be studied. Estragole is an attractor for pollinators and is probably emitted from the flowers.

**Chamber results**

An extensive programme of chamber experiments was carried out in 4 distinct phases over 3 years. Guided by the field measurements, the programme focussed on the photo-oxidation of 7 individual VOCs (α-pinene, β-caryophyllene, limonene, myrcene, linalool, α-terpinene, isoprene) and their mixtures as SOA precursors along with real emissions from tropical plant species.

The first phases were single precursor investigations of the effect of initial concentration on SOA chemical and physical properties, characterising gaseous composition and identifying aerosol components as the particles aged in i) unseeded and ii) seeded photo-oxidation experiments, the latter using both inorganic and oxygenated organic aerosol as seeds. These clearly identified precursor-specific relationships between aerosol composition and properties, discounting generalisation by extrapolation from a single precursor.

The third phase comprised a series of mesocosm experiments (Fig 12 - right) using emissions from three tropical plant species and those from birch species for contrast. The plants were kept under representative temperature, relative humidity and CO	extsubscript{2} conditions and inlet air into the plant enclosure was scrubbed for particles, VOCs and NO	extsubscript{x}. The enclosure exhaust fed the photochemical chamber for photooxidation experiments. It was found (bottom right) that emissions from the tropical fig species mainly comprised isoprene (red), with low monoterpene concentrations (blue), hence forming no measureable SOA. Putative gas phase isoprene SOA precursors (hydroxyacetone and hydroperoxides) were found at very low yields, possibly suppressed by NO	extsubscript{x} levels. In contrast with the tropical species, the birch emissions yielded substantial SOA, owing to the very much higher mono-terpene (largely β-pinene and Δ3-carene) emissions.

Synthetic mixture experiments were carried out in the final phase of experiments to compare the results with those from the mesocosm experiments. The influence of isoprenoid oxidation products on the smog chamber formation of secondary organic aerosol in mixed terpene/isoprene systems was extensively investigated and is the subject of ongoing investigation.

**Mechanism development**

Detailed gas phase mechanisms have been constructed for the monoterpane limonene and sesquiterpene β-caryophyllene, and released as part of the new Master Chemical Mechanism v3.2 (see Figure 13 below).
The boiling points, enthalpies of vapourisation and vapour pressures of all intermediates have been estimated and the gas phase chemistry has thereby been coupled to gas-aerosol partitioning code for prediction of SOA composition for comparison with observations. A detailed mechanism sensitivity appraisal was undertaken to examine processes which might increase the efficiency of the production or recycling of OH radicals during the oxidation of isoprene under low-NOx conditions, and which may therefore help to explain the exceptionally high OH concentrations recently reported over tropical forested regions and their indirect impact on the production of condensable material.

The mechanisms for both limonene and β-caryophyllene have been incorporated into chamber models for prediction of the gas phase evolution and formation and transformation of SOA in the photo-oxidation experiments.

Scale-up modelling

Informed by the chamber evaluated model mechanisms, a series of nested model simulations for the OP3/ACES spatial domain were undertaken. Using a 0.5x0.67 nested grid embedded into the global GEOS-Chem CTM, assimilating NASA GMAO meteorology, predictions of gaseous chemistry and bulk aerosol composition for the ACES / OP3 period were conducted. Using satellite AOD and HCHO products for constraint, budgets and vertical profiles of aerosol properties were simulated, clearly indicating discrepancies between the larger-scale features and more locally influenced in situ airborne and ground-based measurements.
Appraising the Direct Impacts of Aerosol on Climate (the ADIENT consortium)

(Consortium led by Prof. Don Grainger, Oxford University, with colleagues from the Universities of Oxford, Manchester, Reading, Leeds, London Imperial College & STFC-RAL, Lidar Technologies, & Plymouth Marine Laboratory)

Importance of ammonium nitrate and secondary organic aerosol in N. West Europe

Ammonium nitrate and Secondary Organic Aerosol (SOA) were shown to be dominant chemical components of the Northern European sub-micron aerosol burden. Their concentrations across Europe were measured using an Aerosol Mass Spectrometer (AMS) (Fig 15 - right). Their mass fractions ranged from 20-50%. SOA concentrations typically exceeded sulphate loadings across Europe. Ammonium nitrate was found to dominate in North-Western Europe during episodes of high pollution, reflecting the enhanced NO\textsubscript{x} and ammonia sources in this region. Less aged SOA was also observed to peak in this region, indicating rapid formation of SOA in anthropogenically perturbed air masses. The SOA was then observed to age appreciably upon advection downwind. Furthermore, the vertical distribution of both ammonium nitrate and SOA was shown to be highly complex and an important factor in regional climate. In particular, ammonium nitrate concentrations were regularly observed to peak at the top of the boundary layer, where relative humidity is generally highest. Semi-volatile SOA was also often observed to follow the same trend. This greatly enhanced the scattering intensity of the aerosol and led to large increases in aerosol optical depth (see Fig 15, bottom) and associated radiative forcing by close to a factor of two. Such increases have major ramifications for regional climate predictions as semi-volatile components are often not included in aerosol models.

Spatial distribution of black carbon aerosol

The properties and processing of black carbon (BC) aerosol were investigated using a Single Particle Soot Photometer (SP2), flown in the boundary layer across Europe for the first time. This new instrument provided information on the physical properties and the spatial/vertical distribution of BC aerosol in the region. Average BC mass concentrations ranged from about 300 ng m\textsuperscript{-3} near urban areas to approximately 50 ng m\textsuperscript{-3} in remote continental regions, lower than previous surface-based measurements. Black carbon was shown to contribute 0.5-3 % of the sub-micron aerosol mass.

Figure 15: Changes in mass loadings of species (top), of organic (HOA & LV-OOA) mass fractions (middle) as a function of distance from source, and Aerosol Optical Depth (AOD) as a function of nitrate loading (bottom), from all from all ADIENT/EUCAARI flights across Europe
A significant advance was the measurement of BC mass size distributions, which showed that regional BC peaked at approximately 180 nm, while BC closer to source was shifted to smaller diameters (approximately 160 nm). The mixing state of BC was also investigated, with more internally mixed BC dominating further away from major source regions. Such changes in BC mixing state did not appear to lead to an enhancement in aerosol absorption. The single scattering albedo (SSA) was shown to be influenced by both the presence of enhanced BC concentrations and the relative concentration of BC to secondary aerosol components (Fig 16 right), particularly ammonium nitrate and SOA. The SSA was lower in urban plumes, where BC mass concentrations and mass fractions were higher, compared with regional aerosol. These measurements of BC provide much needed constraints for model evaluations of regional and global aerosol impacts. Furthermore, they highlight how the evolution of BC in the atmosphere influences its properties, which determine its impact upon regional climate.

**Characterisation of aerosol optical properties in anthropogenically influenced air masses**

Measurements from the BAe-146 FAAM aircraft were used to characterise the optical properties of aerosol across north-western Europe during May 2008. This period was dominated by a high pressure system over Denmark driving easterly or north easterly flow across much of north-western Europe. Flights were made following the air mass from the southern Baltic Sea to the Atlantic west of Ireland. As the air mass aged, the single scattering albedo tended to increase, and this is consistent with the addition of scattering material resulting from emissions of the precursors of ammonium nitrate aerosol and organic carbon aerosol. Ammonium nitrate was found to be a strong influence on mass extinction co-efficient and single scattering albedo. This has important implications for the modelling of radiative forcing due to anthropogenic aerosol since many models have yet to include this aerosol component.

Enhanced capability of the airborne platform to measure the mass of black carbon aerosol allowed an attempt to obtain closure between measured scattering and absorption and that simulated using measured size distributions and assumed refractive indices. In the case of scattering good agreement was achieved, suggesting that estimates of the real part of the refractive index for aerosol components are well constrained. For absorption, the agreement was not as good.

**Comprehensive analysis of the European aerosol microphysical properties**

Combining ADIENT and EUCAARI airborne and ground-based aerosol measurements from the May 2008 campaign has allowed a complete analysis of the aerosol size distribution over Europe. The primary aim of this analysis was to understand how the uncertainty in the aerosol microphysics is influenced by the poorly constrained properties of the emitted carbonaceous particles. Good consistency was found between the global model (GLOMAP-bin – core project CP3) and observed aerosol size distributions at ground sites in Europe. The agreement is particularly good for particle sizes larger than 100 nm that control the direct forcing, but becomes increasingly uncertain at smaller sizes due to uncertainties in the size of primary particles and nucleation processes.

The model-observation comparison reveals that the size distribution of emitted carbonaceous particles is poorly defined and accounts for much of the uncertainty in the size distribution and cloud condensation nuclei concentrations. However, the effect of the uncertainty on the single-scatter albedo and hence on direct forcing is much less, and not likely to be a major source of uncertainty in the forcing.

**First model evaluation of the black carbon particle size distribution**

Previous model studies as part of AEROCOM analysed the HIPPO trans-Pacific black carbon mass concentrations measured by the SP2 soot photometer. As part of ADIENT
analysis was extended to the black carbon number concentration and size distribution. The observations used were a composite of all FAAM SP2 measurements in the European boundary layer during May 2008. The comparison showed that the total concentration of modelled BC particles in the observed size range (~50-200 nm) was very good. The total mass was also within a factor of two. However, the modelled size distributions over predicted concentrations at small sizes and under predicted at larger sizes. Such a discrepancy was not apparent in the all-component size distributions. This difference may arise because the distribution of BC in the emitted carbonaceous particles is different to that assumed in all models (including GLOMAP), where BC is ascribed a fixed mass ratio in all emitted sizes. This assumption may need revisiting as there may be significant differences in SSA due to the different modelled and observed BC size distributions. Should the distribution of BC not be uniform in the emitted carbonaceous particles, it will prove difficult to handle a more complex assumption in the modal models used in climate assessments.

**Testing the importance of model complexity**

A key question in ADIENT was whether a more complex representation of the aerosol microphysics would lead to better predictions of aerosol in polluted European conditions. The GLOMAP-bin model is an example of a complex model, while GLOMAP-mode is an example of a simpler model that is also used in the HadGEM-UKCA composition-climate model (CP3). Therefore, these two models were compared to determine whether there was observational evidence for better performance of the more complex model. The interaction of emitted carbonaceous particles with existing particles was compared by having either one distribution for all components, or by treating BC particles separately (which adds cost to the model). The conclusion is that, while there are differences between bin and modal treatments, the differences (see Fig 17 below, bin model AOD left and Modal model AOD right) are not significant when compared to observations. Most importantly for ADIENT, the AOD in the bin and modal models is probably the least sensitive to microphysical complexity. We therefore conclude that the UKCA modal model is sufficiently complex in terms of its representation of aerosol size distribution to make reliable predictions of AOD and direct forcing.

![Figure 17; GLOMAP AOD using (a) GLOMAP-Bin model (left) and GLOMAP-mode model (right)](image)

**Testing methods to derive top of the atmosphere reflected shortwave fluxes in the presence of aerosol during the ADIENT campaign: application to GERB and SEVIRI**

Due to their fifteen minute temporal resolution, the combination of the narrowband Spinning Enhanced Visible and InfraRed Imager (SEVIRI) and the Geostationary Earth Radiation Budget (GERB) instruments on the Meteosat Second Generation series of satellites offers an opportunity to track the evolution of aerosol and assess their associated radiative impact over the approximate 60°S-60°N, 60°E-60°W region viewed by the instruments. However, the aerosol retrieval algorithm used to generate optical depths in the standard GERB products uses one fixed aerosol model which may not be appropriate for specific aerosol events. In addition, fluxes are typically derived from satellite observed radiances via the application of an angular distribution model (ADM) selected according to scene type. At present, shortwave (SW) fluxes derived from the observed GERB radiances do not contain a treatment to account for the impact of aerosol on the anisotropy of the radiance field, an omission which will result in angle dependent SW flux biases.

Two methodologies to correct for these SW flux biases were evaluated. The first involves the application of a theoretical correction to clear-sky ADMs derived from the Clouds and the
Earth's Radiant Energy System (CERES) instrument to account for the presence of aerosol. The second uses CERES observations, in this case building ADMs as an explicit function of near surface wind speed and aerosol optical depth. The analysis focused on periods and locations sampled during the ADIENT/EUCAARI projects. Both methods gave a significant reduction in the instantaneous direct radiative efficiency of aerosol calculated using the GERB observations as they stand, and after application of the new ADMs. By considering the homogeneity of the derived fluxes as a function of solar and viewing geometry it was concluded that for the conditions sampled during the flights, the first aerosol correction method gave the most reliable results.

Research then focused on a specific flight off SE England during May 2008 to assess whether radiative closure could be achieved using the recommended flux conversion and the observed aircraft data. Whilst a limited closure was possible, the aerosol optical depths retrieved using aerosol optical properties inferred from the aircraft tended to be somewhat smaller than those recorded by nearby Aerosol Robotic Network (AERONET) stations. Better agreement on this wider spatial scale was obtained using the standard fixed aerosol model. The results highlighted the effect of small scale inhomogeneities in aerosol composition on satellite derived aerosol fields, and the implications for calculations of their associated direct radiative effect.

**Comparisons of model radiance simulations with global satellite measurements**

Traditional satellite - model aerosol comparisons have focused on comparing satellite derived aerosol optical depths directly with those predicted by models. These comparisons are valuable but may be limited by potentially inconsistent assumptions concerning the optical properties of aerosol. An alternative is to compare radiances predicted from model aerosol fields with the directly observed satellite radiances. Radiances from the ATSR series of satellites were used for the comparison as the instrument has a long measurement period (1995-present), dual view and excellent calibration.

The model and satellite see some of the same large-scale features in the aerosol distribution. However AATSR detects more aerosol in the Saharan dust region and northern latitudes than GLOMAP. In the southern hemisphere the model and satellite are in broad agreement with the satellite possibly observing fewer aerosols in some regions. Comparisons of GLOMAP and AATSR optical depths and radiances show significantly lower amounts of aerosol and consequently produce significantly lower top of atmosphere radiance than observed by AATSR. This may in part be caused by cloud-contamination in the satellite radiances, but it seems likely that there is a general underestimation of either aerosol amount or its radiative effect by the model. Further work to diagnose this discrepancy and improve the level of agreement would clearly be valuable.

**Estimation of radiative forcing due to anthropogenic aerosol**

A detailed regional analysis of the GlobAEROSOL AATSR data was undertaken, including evaluation against AERONET. This produced a bias-corrected, error-characterised monthly/regional aerosol optical depth dataset. The Edwards and Slingo radiation code has been used with the GlobAerosol satellite data set to estimate the seasonal variation of regional mean radiative forcings from 2006 compared to pre-industrial levels of aerosol. The global and annual mean clear sky radiative forcing is -0.15 Wm\(^{-2}\) at the top of the atmosphere and -3.1 Wm\(^{-2}\) at the surface, which is in line with previous studies. Significant error sources in the data and radiative transfer calculations have been characterised and propagated through to the resulting radiative forcing. Uncertainty caused by the possible errors in the surface albedo and by neglecting spectral variability of aerosol optical properties is small (less than 1 %) compared to that caused by spatial and temporal averaging of the AOD observations and assumptions about scattering properties (up to 10 % in each case). Also there is little to be gained from using high spatial or temporal data on aerosol optical depth unless the aerosol scattering properties are also known with high spatial and temporal resolution. This suggests that satellite retrievals of aerosol properties such as single scattering albedo, systematic ground based or airborne based monitoring of aerosol properties, or the use of extensively validated aerosol transport models are required before a reduction in uncertainty in aerosol forcing can be made.
**Aerosol Interactions in Mixed Phase Clouds (APPRAISE-Clouds consortium)**

(Consortium led by Prof. Tom Choularton, University of Manchester, with colleagues from the Universities of Manchester, Reading, Leeds, York, Hertfordshire and London Imperial College)

Work within this consortium included laboratory studies on the ice nucleating properties of dust and soil aerosol, principally carried out at the AIDA chamber at Karlsruhe Institute of Technology (KIT) in Germany (Fig 18). This was supplemented by further microphysical studies of the aggregation and growth of ice crystals in the Manchester Ice Cloud Chamber, MICC (Fig 19). In addition to these fundamental laboratory studies, extensive in situ measurements of aerosol and cloud microphysics were made from an aircraft. These were carried out in conjunction with simultaneous remote sensing measurements and comprehensive ground based measurements. Frontal, stratiform and convective clouds were investigated. Such measurements were performed with a view to better understanding cloud aerosol interactions in mixed phase clouds. Models developed as part of the core project CP4 were used to link the chamber studies to the field observations and facilitate their interpretation.

The remote sensing data were principally obtained using the Chilbolton Facility for Atmospheric and Radio Research (CFARR), in southern England (Fig. 20). Range Height Indicator (RHI) scans from the steerable 3 GHz dual polarisation Doppler Radar, CAMRa (Chilbolton Advanced Meteorological Radar) were conducted along a radial where the aircraft was flying and obtaining concurrent in-situ measurements of the cloud microphysical properties. The UK Facility for Airborne Atmospheric Measurement (FAAM) BAe146 aircraft was used for this purpose (Fig. 20). Other measurements from the CFARR site included those from the vertical pointing 35 GHz Doppler cloud radar (Copernicus) and those from various lidars.

In total, APPRAISE-Clouds flew 28 science flights, of which 21 involved flying in the vicinity of CFARR. Chosen case studies were simulated using the Advanced Research WRF (Weather Research and Forecasting) model version 3.1, generally employing the dual moment Morrison microphysics scheme which predicts the mixing ratios and number concentrations of hydrometeors in 5 categories (droplets, rain, ice, snow and graupel). WRF simulations were run with 4 nested domains from 27 km down to 1 km horizontal resolution (in a 3:1 ratio) and initialised/driven at boundaries by GFS data.
Detailed modelling of the microphysics and dynamics of individual case studies was also undertaken using various forms of the ACPIM model developed within CP4.

**Main Findings**

Results from the chamber studies have shown that dust nucleation is much more efficient in the condensation freezing mode, consistent with observations in cloud. In this mode, many dust and soil particles are found to operate as ice nuclei at temperatures below -12°C, with an increasing number being active at decreasing temperature. These measurements also suggest that ice nucleation is a singular process i.e. a particular particle will always act as an ice nucleus in the same conditions and this is not time dependant. This is consistent with much of the APPRAISE-Clouds field observations for temperatures below about -20°C. For layer clouds with tops warmer than this (and indeed as warm as -8°C) ice nucleation has been seen to produce very low concentrations of ice crystals (often less than 0.1L⁻¹). However in one case (Fig 21 below), a shallow layer cloud with cloud top (CT) temperatures greater than -13°C was able to persist for many hours maintaining an almost constant concentration of ice, even though the crystals grew to form snow which fell and precipitated out of the cloud. Since crystal concentrations were maintained, this could suggest a stochastic nucleation process was in operation. An alternative explanation for the replenishment of crystal numbers is that contact nucleation from interstitial particles diffusing to water droplets was occurring over time.

![Figure 21: Copernicus (vertical pointing) radar data showing a shallow layer cloud overlying CFARR from 06:00. In situ aircraft observations observed a CT temperature of -13°C, with liquid water at CT and low concentrations of ice throughout. The cloud persisted for over 24 hours even though it was gently precipitating snow through its base. By contrast, an embedded convective cell (seen passing over CFARR at 07:15) was seen to contain significantly higher concentrations of ice than in the layer cloud above.](image)

A statistical analysis of 4 years of lidar & radar observations from CFARR of cloud layers containing ice was undertaken to derive the fraction of such clouds that also had supercooled liquid water present at cloud top. The results (Fig. 22) indicate that most ice clouds have some liquid at CT when CT temperatures are warmer than ~ -20°C; about half have liquid water present at CT when temperatures are between -20°C and -27°C; while none have liquid present at temperatures of -37°C or below. The field measurements suggests that for temperatures warmer than -27°C ice is formed by heterogeneous nucleation of the cloud droplets present. At temperatures lower than this, ice is formed by a mixture of homogeneous and heterogeneous nucleation at CT. Measurements from SID-3 (a new Small Ice Detector instrument further developed as part of the APPRAISE-Clouds project to detect the size and shape of small ice crystals) have revealed that these ice crystals are often very rough. This new finding is important for modelling the optical properties of the ice particularly with respect to the scattering of short wave radiation.
Frontal clouds

Nucleation at cloud top is seen as the primary source of ice in deep frontal clouds. The ice nucleated at cloud top (e.g. Fig 23, top) grows via deposition/aggregation into snow (Fig 23, middle). Pristine plate-like ice crystals are sometimes observed (at temperatures around -20°C) in regions of high differential reflectivity (as identified by the CAMRa). Embedded convection originating from near the surface is frequently observed too, and is associated with the production of regions of supercooled water within the frontal cloud. High number concentrations (peaking at over 100 L⁻¹, but typically ~40 L⁻¹) of ice crystals are often found in the temperature range -3°C to -8°C. These ice crystals are relatively small (<300 µm in length compared to over 1 mm for snowflakes) and appear as columns (Fig 23, bottom). The high ice crystal concentrations are most likely a result of Secondary Ice Production (SIP) resulting from the Hallett-Mossop (HM) process of rime splinter ejection during riming. The horizontal extent to which SIP appeared to influence ice number concentrations was over 10's of km in many cases, and these regions generally contained a significant amount of condensate (peaking at over 0.5 g m⁻³). WRF simulations of these cases showed the model was unable to reproduce the observed number of SIP particles associated with snow falling through the HM zone. However, the precipitation distribution and amount was sensitive to the treatment of the HM process in the model. This process results in the modification of the precipitation distribution within the system and is responsible for regions of intense precipitation without altering the total precipitation from the system.

Convective Clouds

In the convective cloud cases investigated, ice was seen to be initiated heterogeneously near CT in small concentrations, and so secondary ice particle production by the HM process was the dominant mechanism for controlling ice crystal number. These crystal numbers were well reproduced by WRF and by simulations using the core model, and were associated with the riming of graupel particles falling through the HM zone. Further simulations with the core model, using ground based and airborne aerosol measurements as input, were able to elucidate the detail of the dynamical structure of these clouds, the cloud droplet and ice nucleation and the partitioning of aerosol, notably black carbon between the cloud droplets and interstitial air.

For one case study where a line of shallow convective clouds was observed, the WRF model was used to assess the ability of the HM process to control precipitation formation (where CT temperatures were no colder than -8°C). The results indicated that while HM does act to increase the mass and number concentration of ice produced in these model simulations, in the absence of HM the ice number concentration arising from primary ice nucleation alone (several per litre) was apparently sufficient to sustain precipitation. Thus in the model, at least for this particular case, the Hallet-Mossop process was shown to be non-critical for the formation of precipitation, although it did alter the extent and shape of the precipitation field.
4. APPRAISE Knowledge Exchange Activities

Knowledge exchange (KE) activities during the APPRAISE programme were organised in the first instance through UKAAN, the UK Atmospheric Aerosols Network. UKAAN developed and maintained a website at the University of Reading which provided a wealth of information about aerosols and aerosol related subjects from the past, present and future. This information included: the latest aerosol and aerosol related news; links to datasets (both aerosol and aerosol related) including those from both measurement and modelling activities; information about aerosols by aerosol type and event; aerosol field experiments and other case studies involving aerosol; aerosol related organisations; aerosol meetings, and aerosol jobs. UKAAN produced monthly newsletters disseminating the latest information in each of these areas. Contact details of aerosol scientists and their specialisations, updates on dataset availability, information on aerosol instrumentation and aerosol models, and lists of published papers were all available through this close coordination between APPRAISE and UKAAN. The co-location of a wide variety of aerosol information in this way alongside the activities within APPRAISE enabled the formation and development of a true aerosol research community. Following the end of UKAAN activities in spring 2008, much of this information is now held on the NCAS Composition or NCAS APPRAISE websites.

During 2008 it was decided to add the remaining APPRAISE KE funding allocation to the larger pot of KE funding that was available through the NERC July 2008 KE funding round. This was undertaken so as to maximise the funding available to prospective APPRAISE KE projects as the programme was coming to a close. One APPRAISE earmarked proposal was funded through this route. This project was managed by Phillip Stier from the University of Oxford, who organised the 9th AEROCOM workshop which was held in Oxford in 2010 effectively with financial support from the APPRAISE KE budget. The AEROCOM project is an open international initiative of scientists from around the world interested in the advancement of the understanding of the global aerosol and its impact on climate. To this end, a large number of observations (including satellite and surface site data) and results from more than 14 global models have been assembled to document and to compare the “state-of-the-art” modelling of global aerosols. Within AEROCOM models use a common protocol along with standardised input (e.g. the AEROCOM emission inventories) so as to aid comparison. Results are then documented via interactive websites which provide access to 2-D fields and standard comparisons to observations. Regular workshops (like the 9th hosted in Oxford in 2010) then discuss findings and future directions. These workshops thus provide a forum for the use and comparison of field and remote observations, common emission inventories and global models for comparison and assessment of progress of understanding of global aerosols and their effects on climate.

These KE activities stand alongside those which occurred daily throughout the period of APPRAISE. Each APPRAISE project introduced a number of new young investigators to the field of aerosol research, training them to become scientists and experts in the field. In addition, information was also been disseminated to the general via public meetings and particularly through interviews with the media. There have been a number of occasions where APPRAISE researchers have been interviewed by reporters from the local, national or international press, radio and television. In this way, contributions have been made to general interest newspaper articles or radio/television programmes (e.g. the BBC One show) or to more scientific publications or programmes (e.g. TV programmes about the environment, weather and/or climate, e.g. as shown on the BBC and Discovery channels).

5. The APPRAISE legacy and Aerosol Research post- APPRAISE

One of the lasting legacies of the APPRAISE programme is the strength and depth of the UK aerosol research community that has been built up as a result of the programme. As discussed earlier, (section 2 - Generic Achievements), prior to APPRAISE no such community existed. Now, however, this community is comprised of laboratory and fieldwork scientists, instrument scientists and computer modellers (working over a range of scales from that of individual aerosols up to global scales), and includes a range of new young
investigators introduced to aerosol science and trained to be experts in the field during APPRAISE. Together, this community is now equipped to take on remaining and new challenges in the field of aerosol science.

In generating this UK aerosol community, not only have links been created between research groups in a variety of disciplines within the UK, but this has also been extended internationally, particularly across Europe. This was achieved by collaborating with projects that had similar goals, such as the EU framework 6 Programme, EUCARI, or the international AEROCOM project. The EUCAARI co-ordinator, Prof. Markku Kulmala from Helsinki, made a keynote presentation at the APPRAISE kick-off science meeting (hosted by UKAAN at Reading – see section 4). The APPRAISE Steering Group Chairman (and later the Programme Advisory Group chairman), Urs Baltensperger, was also a work package manager within EUCAARI. He made a keynote presentation at the final APPRAISE science meeting (in Exeter in 2011). EUCAARI’s objectives were to investigate the role of aerosol on climate and air quality, goals which were completely complementary to those of APPRAISE. However, by working together much more was achieved than was possible by working apart. A perfect example of this was the collaboration between ADIENT and EUCAARI in the EUCAARI-Longrex experiment, looking at the formation and aging of aerosols as they advected across NW Europe in May 2008. By deploying two aircraft, one (the DLR Falcon) working at high levels remotely sensing the aerosols in the vertical column below, enabled the FAAM BAe146 aircraft to fly in situ and sample aerosol layers so thin they would be almost impossible to locate (or to even know of their existence) without such collaboration. However, by working together and sharing resources, data were obtained which would otherwise have been missed by both projects (e.g. the simultaneous remote sensing and in situ collection of data). These links were further strengthened by the sharing of raw data, analyses and by presenting at each others science meetings and workshops. This led to joint publications in peer reviewed journals and presentations at international conferences. Links like these, forged throughout the APPRAISE programme are another of its legacies.

International links were also created by inviting other world leading experts in aerosol or cloud disciplines to present keynote presentations at the annual APPRAISE science meetings. Not only did this provide an opportunity to advertise the work of APPRAISE to key players in the international community, it also enabled APPRAISE output to be placed within an international context. This not only benefited the APPRAISE programme, but also led to continued or new collaborations between the international community and UK projects and programmes outside of APPRAISE. Other invited keynote talks were given by members of the APPRAISE end-user community; often these were investigators in key end-user areas at the Met Office.

In addition to building a community of researchers, APPRAISE has also built up an unprecedented and varied data set throughout its existence. These data from: measurements obtained in field experiments from as far afield as the tropical rainforest in Borneo to the skies over north-western Europe; laboratory studies examining the properties of individual aerosols trapped in optical tweezers or electrodynamic balances to those being studied in aerosol environment chambers or cold cloud chambers; output from models of varying complexity over a range of scales from process level up to global climate model scale. All of these data are archived at the British Atmospheric Data Centre (BADC), or have links to them if they are stored at other locations outside of BADC. This includes access to data from outside the APPRAISE programme, obtained through the close links forged throughout APPRAISE as described above. Thus, the APPRAISE data set is a true legacy, and is available to current and future aerosol research scientists in the UK and across the world.

Another legacy of APPRAISE is the array of analysis techniques and instrumentation developed within the programme which are now available to the community. These include analysis and separation techniques developed in core project CP1b to determine the composition and/or chemical groupings present in samples such as those collected in the rain forest or laboratory aerosol chamber in ACES. This includes the development of a comprehensive two dimensional gas chromatography and mass spectrometry (GCXGC-TOF/MS) technique to look at volatile and semi-volatile polar compounds in SOA, and the
development of high performance liquid chromatography coupled with tandem mass spectrometry (i.e. MS-MS) techniques to resolve the molecular structures of samples (e.g. of oligomeric compounds in SOA formed via heterogeneous reactions of monomers). The method of trapping individual aerosol particles, either in an optical tweezer arrangement or in an electrodynamic balance, both developed within core project CP1a, proved to be powerful techniques for examining the uptake of water to or reactivity of aerosol particles.

The development and use of instrumentation to measure the properties of ice particles, either in-situ in cloud or in the ice-cloud chamber, is another legacy of the programme. Included in this is the development of the Small Ice Detector, SID-3 (by the University of Hertfordshire) within APPRAISE-Clouds, to the point where it is now able to see the roughness of ice particles which is crucial to calculating the light scattering properties of these cloud particles. Also the development of software to analyse the images from modern fast 2-D imaging cloud probes like the SPEC 2DS and DMT CIP probes is another legacy. This software was primarily developed within APPRAISE-Clouds to analyse the measurements obtained within the project. However, this software will be (and in fact already is) extremely useful to others in the community that use such probes in aircraft and laboratory investigations of clouds and cloud properties.

Another major legacy of the APPRAISE programme is the development of an array of models and modeling tools. Many of these were developed initially within the APPRAISE core projects, and then used extensively within the consortium project work. However, these are available now for use in projects following on from APPRAISE. As an example, the tools and hierarchy of radiative transfer models (developed in CP2), including the RFM DISTORT benchmark radiative transfer model and the aerosols optical property database (see section 3 highlights), continue to be used to help develop and test GCM radiation schemes (e.g. within AEROCOM). They are also being developed further to expand their scattering framework to include non-spherical scattering (e.g. of desert dusts as in the current NERC FENNEC consortium project). Similarly, the family of aerosol models produced in core project CP3, to help to develop the inclusion of aerosol processes in Global Climate Models, and to determine the level of complexity required, led to the inclusion of the GLOMAP-Mode aerosol module in UKCA and the HadGEM3 Met Office Climate Model. This model, and the hierarchy of models of increasing complexity used to produce it, are now available to be used and further developed in post APPRAISE studies (as is already happening). In a similar way the model framework developed in core project CP4 to understand the detailed aerosol processes and their impact on clouds subsequently forming on them, used extensively in APPRAISE-Clouds is to be a cornerstone of work which follows on in the NERC Aerosols and Clouds research project.

Finally, one of the most important legacies of the APPRAISE programme is its list of publications, since this represents the main scientific output of the programme. This list includes a significant number of high quality peer reviewed papers produced throughout the lifetime of APPRAISE, as well as a collection of the papers that have been presented at an array of national and international conferences and workshops by APPRAISE investigators. A list of these publications was initially started as a result of the coordination of APPRAISE KE activities through UKAAN, but the list (of peer reviewed publications) is now is to be maintained and continually updated on the NCAS APPRAISE website (see also Appendix 1).

It can be seen from this report that the APPRAISE programme has a significant list of achievements and leaves a legacy which is already being taken advantage of in the research which is continuing now that APPRAISE has come to an end. Many members of the aerosol research community built up within APPRAISE are heavily involved in the research being undertaken in successor projects and programmes such as the NERC AEROS project and the “Aerosols and Clouds” research programme. The latter is effectively the successor programme to APPRAISE, and seeks to build on much of the scientific progress achieved within APPRAISE, by concentrating on a programme of laboratory investigations, modelling and model development. These were considered areas needing further investigation post-APPRAISE to reduce the remaining uncertainties in our understanding of aerosols and clouds, and their role in the atmospheric energy budget and future climate change.
Appendix 1: APPRAISE Peer Reviewed Papers


Gabey A. M., W. R. Stanley, M. W. Gallagher and P. H. Kaye. 2011. The fluorescence properties of aerosol larger than 0.8 µm in urban and tropical rainforest locations. Atmospheric Chemistry and Physics. (11), 5491-5504. ISSN: 1680-7316. DOI: 10.5194/acp-11-5491-2011


Pope F.D., B.J. Dennis-Smithter, P.T. Griffiths, S.L. Clegg, R.A. Cox. 2010. Laboratory and modelling studies of aerosols comprised of malonic acid, glutaric acid, and their mixtures with sodium...
chloride: Part I hygroscopic growth. J. Physical Chemistry A. 114(16), pp 5335–5341. DOI: 10.1021/jp100059k


Sayer A. M., G. E. Thomas, P. I. Palmer and R. G. Grainger. 2010. Some implications of sampling choices on comparisons between satellite and model aerosol optical depth fields Atmospheric Chemistry and Physics, 10(22), pp 10705-107. ISSN: 1680-7316. DOI: 10.5194/acp-10-10705-2010


