



IGAC *tivities*

Newsletter

of the International Global Atmospheric Chemistry Project

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A Note from the IGAC Co-chairs: Sandro Fuzzi and Phil Rasch

The 9th IGAC Science Conference, co-sponsored with CACGP and WMO, and held in Cape Town, South Africa, September 17-22, 2006 is now behind us. This IGAC key event was extremely well attended by 410 participants, coming from 48 countries and was centered around the overarching subject "Atmospheric Chemistry at the Interfaces". The eleven sessions organized within the Conference addressed the issue of interfaces at different levels: between phases in the atmosphere, between compartments of the Earth System and between disciplinary approaches – each of which emphasized the complexity of the Earth System and the need for a truly interdisciplinary approach to studying it.

Three of these sessions were jointly organized with the IGAC partner projects iLEAPS, SOLAS and WCRP-SPARC. Three further "mini-sessions" were devoted to programs of specific interest to the African continent (AMMA, DEBITS and APINA). This was the first IGAC Conference held in Africa and it was particularly rewarding to see the unusually large participation of African scientists (77 total coming from 10 countries). In fact, among the goals of the IGAC Conferences is to promote the engagement of scientists from emerging countries. The African continent is very important for global change science both in terms of relevant emissions and processes and in terms of effects. We still know far too little about the environment in this continent, and there is a pressing need for an increasing engagement of local scientists in the field of global change science in general and in atmospheric chemistry in particular. As noted in one of the articles herein, there is also a need to better link communities studying atmospheric chemistry for climate and those studying it in the context of air pollution. To this end, we can certainly say that this Conference was an important step forward in this direction. The participation of young scientists, another relevant aim of IGAC, was also high and the attendance of 46 young scientists from 23 countries was favored through travel grants awarded by the three co-sponsoring programs, the European ACCENT project, NASA and the University of Witwatersrand in South Africa.

We would like to thank Mark Lawrence and the International Programme Committee and Stuart Piketh and the Local Organising Committee for organizing this extremely successful event, and Barry Huebert, the keynote speaker of the Conference who gave an inspiring introduction on the interfaces between science, society, environment and man-driven global changes.

A second important activity that IGAC has been involved in over the past months was the fostering of the Atmospheric Chemistry & Climate Initiative, a joint effort of IGBP-IGAC and WCRP-SPARC. The effort was initiated in March 2006 with the endorsement of WCRP and IGBP. At an initial meeting in August a small group gathered to outline the goals of this initiative, which will focus on improving process representation in chemistry-climate models. The first Atmospheric Chemistry and Climate open workshop will be held in January 2007 with the aims of engaging key modeling communities in this effort and of establishing a more concrete implementation plan. This effort will build on and be coordinated with the work ongoing under the CCM-Val (stratospheric Chemistry Climate Model Validation), AeroCom (Aerosol Model Intercomparison Project) and ACCENT-MIP (ACCENT Model Intercomparison Project) activities.

This issue of IGAC *tivities* is comprised of a few contributions that illustrate the many activities of the European Network of Excellence ACCENT on the subject of Atmospheric Composition Change. The objectives of this large European Program are strongly overlapping with those of IGAC and, in fact, one of the aims of ACCENT is to facilitate the European contribution to IGAC activities, also through supporting the European IGAC Office.

The last contribution to this issue concerns a workshop that was held joint by IGAC WCRP's SPARC and GEWEX projects on "Processes Controlling the Chemical Composition of the Tropical Tropopause Layer". Consistent with the theme of this years' conference, this workshop was an important step in the direction of bringing together researchers in atmospheric chemistry, dynamics and cloud processes.

P.S. This issue represents the last that Sandro Fuzzi will oversee as an IGAC co-Chair, as his appointment finishes with the close of 2006. We (PJR and SJD) and the IGAC SSC would like to thank Sandro for the energy and attention he has put into IGAC over the last three years. It has been a pleasure to work with him and we wish him enjoyment and productivity in his next endeavors.
~Phil Rasch, co-Chair & Sarah Doherty, Executive Officer

Science Features

ACCENT: The European Network of Excellence on Atmospheric Composition Change

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Changes in atmospheric composition directly affect many aspects of life, determining climate, air quality and atmospheric inputs to ecosystems. In turn, these changes affect the fundamental necessities of human existence: human health, food production, ecosystem health and water. Atmospheric composition change research is therefore fundamental to the future orientation of the world's sustainable development strategy. To deal with the aforementioned environmental problems, the Network of Excellence ACCENT was established by the European Commission within the 6th Framework Program for Research and Technological Development with the overall aim of promoting a common European strategy for research on atmospheric composition sustainability, developing and maintaining durable means of communication and collaboration within the European scientific community, interacting with the international scientific community, facilitating research in the field, and optimizing interactions with policy-makers and the general public.

The Network includes 41 European Institutions from 21 Countries, one International Organisation (IIASA), and the Joint Research Centre of the European Commission. The Partners involved in ACCENT (<http://www.accent-network.org/portal/project-description/participant-list>) represent points of excellence in the field of atmospheric composition change but, in order for ACCENT to succeed in the planned integration and promotion of research on atmospheric sustainability in Europe, a much larger scientific community needs to be involved in the activities of the Network. For this reason, a second level of participation within ACCENT was built, consisting of a much larger group of Associates who can contribute to ACCENT and, at the same time, benefit from the co-ordination and the activities that ACCENT undertakes (conferences, workshops, access to data, access to facilities, web-based information, education and training, etc.). After the first two years of the project, 124 Institutions from 33 Countries, including extra-European Countries

such as the U.S., Canada, Japan, and China are now ACCENT Associates (<http://www.accent-network.org/portal/project-description/associate-list>) and are actively contributing to the activities of the Network.

The overall goals of ACCENT are pursued through specific activities which can be grouped as:

- 1. Joint research program.** A broad common research agenda agreed by the Partners in the Network, also in collaboration with the wider research community, is the basis for a real integration of the European research efforts in this field, and for linking national programs to joint European and international research Projects.
- 2. Tasks for integration.** ACCENT provides a framework for co-ordination and communication among the Partners in the Network and the wider research community. This will be accomplished through a number of tasks organised by ACCENT.
- 3. Outreach tasks.** ACCENT involves European institutions and scientists at the highest level of excellence in the field of atmospheric composition research. However, in order to reach its overall objectives, ACCENT must reach out to the whole of Europe's scientific community and raise the standards of European research. An important aspect of this is the fostering of new expertise, in particular in the area of sustainability research, and the creation of interactive links with policy and the public.

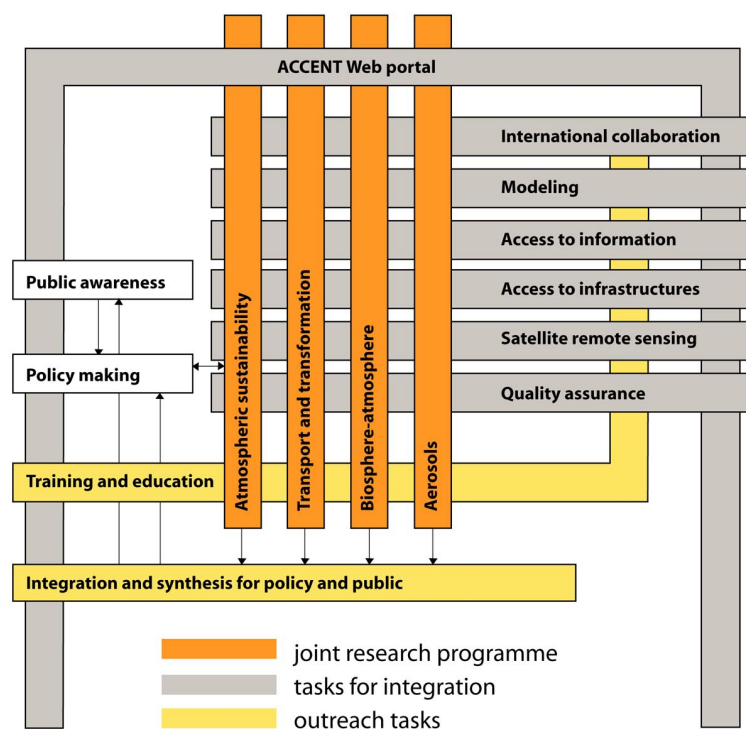


Figure 1. Structure of the ACCENT joint program of activities.

A diagram representing the structure of ACCENT is given in Figure 1. The joint research program is comprised of four vertical subprojects dealing with i) Aerosols, Air Quality and Climate; ii) Biosphere-Atmosphere Exchange of Pollutants; iii) Transport and Transformation of Pollutants; and iv) Atmospheric Sustainability. The joint research program is carried out by means of tools made available through the horizontal tasks: i) International collaboration; ii) Modelling; iii) Access to information; iv) Access to infrastructures; v) Satellite remote sensing; and vi) Quality assurance of atmospheric measurements. The two outreach tasks: "Training and Education" and "Integration and Synthesis for policy making and public awareness", which are strictly connected to both the joint research program and the tasks for integration, represent the societal link of ACCENT.

As can be seen, the objectives and the research topics of ACCENT are overlapping with those of IGAC, and, in fact, the two projects have already collaborated organizing joint workshops on key scientific issues important for atmospheric chemistry and global change (Atmospheric Organic Aerosol, Aerosol Indirect Effect, UTLS Processes, just to name a few), aiming at setting priorities for future research. In fact, one particular aim of ACCENT is to facilitate the contribution of European research groups to IGAC activities. In this respect, a notable example is the active involvement of ACCENT groups in the newly-launched IGAC/SPARC initiative on Atmospheric Chemistry and Climate.

The five contributions which follow this short introduction are derived from ACCENT activities and are just an example of the many initiatives, both scientific and outreach, carried out by the Network.

All activities within the Network benefit from a professionally designed web portal (www.accent-network.org), which is intended to maintain communication within the ACCENT consortium, disseminate knowledge to users outside the Network, and provide a platform for dialogue between research and society.

After almost three years of operation, ACCENT has become a worldwide recognised and appreciated European program and collaborations have been established with programs and organizations worldwide. Besides supporting the European IGAC Office, which facilitates the European contribution to a series of IGAC and IGBP activities, ACCENT contributes to the IPCC reports, WMO-IGACO (World Meteorological Organization - International Global Atmospheric Chemistry Observations), and EMEP (Evaluation of the Long-range Transmission of Air Pollutants in Europe; www.emep.int).

The first ACCENT Symposium (Urbino, Italy, 12-16 September 2005) "The Changing Chemical Climate of the Atmosphere" was a major event which allowed the whole ACCENT community (Partners and Associates) to review the latest scientific progress on the changing atmospheric composition, to provide a "trade-fair" for the various ACCENT activities aimed at integrating atmospheric composition and chemistry research in

Europe, and offered an opportunity for interaction among scientists, policy makers and general public.

A second ACCENT Symposium "Atmospheric Composition Change: Causes and Consequences - Local to Global" is presently being organized again in Urbino, to take place 23-27 July 2007 (<http://www.accent-network.org/2nd%2Dsymposium/>).



Understanding and Quantifying the Atmospheric Nitrogen Cycle

Conclusions from the second ACCENT Barnsdale T&TP Workshop

Contributed by Paul S. Monks Uni-Leicester, UK (P.S.Monks@leicester.ac.uk), Ronald Cohen Uni-California, Berkley, USA, Matt Evans Uni-Leeds, UK, Mike Jenkin, Imperial College, UK, Dave Johnson Uni-Reading, UK, & Peter Borrell P&PMB Consultants, Newcastle-under-Lyme, UK

Nitrogen compounds play a central role in air pollution. They are, in general, pollutants themselves and many play a crucial role in the processing of other trace substances in the atmosphere to form further secondary pollutants such as ozone. Fixed nitrogen is directly involved in photo-oxidant formation in the atmosphere, acidification of ecosystems; eutrophication of surface waters and nitrogen input to soils. To this must be added the formation of aerosols by which nitrogen may be transported to previously pristine regions far from the pollutant sources.

While the central role of nitrogen in the environment is fully appreciated, the details of the formation of many nitrogen compounds, their circulation and transport in the atmosphere, and the mechanisms of deposition and reaction on plant and other surfaces is not as well characterised or, in some cases, well-understood. It is for this reason that the ACCENT Barnsdale workshop on fixed nitrogen was held under the auspices of ACCENT (Transport and Transformation of Pollutants) T&TP sub-project.

ACCENT T&TP is one of the four components of the ACCENT (Atmospheric Composition Change: a European Network) jointly executed research program. Its aim is to bring together and coordinate the European community of researchers concerned with atmospheric chemistry in order to pinpoint current key issues, and to foster research work aimed at resolving the principal areas of uncertainty, so that the models used for analysis and forecasting on global, regional and local scales are more precise and reliable.

The ACCENT T&TP workshop was intended to identify

and review the key uncertainties in the land-atmosphere, atmospheric and laboratory data for nitrogen cycling and to provide a basis for future collaborative research priorities in this area. There were three topic areas addressed:

1. The global atmospheric cycle for fixed nitrogen.
2. Uncertainties in the atmospheric chemical processing of nitrogen compounds.
3. Surface – atmosphere exchange of fixed nitrogen.

A full account of the workshop, including all the individual contributions and the detailed references, is available in the ACCENT report of the workshop (Cox et al., 2006).

Here we shall report on the first two topics; the third is dealt with in an accompanying article by David Fowler and his colleagues (Fowler et al., 2006).

The Global Atmospheric Cycle for Fixed Nitrogen

The global atmospheric cycle for fixed nitrogen can be represented by the processes describing emissions, chemical transformations, transport and removal. Several reviews of the subject are available (e.g Galloway et al., 2004). By way of definition NO_x is $\text{NO} + \text{NO}_2$ and is also sometimes referred to as “active nitrogen”. The sum of total reactive nitrogen or total odd nitrogen is often referred to as NO_y and can be defined as $\text{NO}_y = \text{NO}_x + \text{NO}_3 + 2\text{N}_2\text{O}_5 + \text{HNO}_3 + \text{HONO} + \text{PAN} + \text{MPAN} + \text{nitrate} + \text{alkyl nitrate}$. NO_y can also be thought of as NO_x plus all the compounds that are products of the atmospheric oxidation of NO_x . NO_z is $\text{NO}_y - \text{NO}_x$. The meeting was comprised of highlights of recent progress and a discussion of open questions.

Emissions

NO_x emissions in low NO_x environments. Research interpreting satellite observations of NO_2 shows it is possible to translate observations of column concentrations into emissions and thus provide global information on NO_x sources. For example, Jaegle et al. (2005) suggest that soil emissions may be underestimated by a factor of two. Bertram et al. (2005) show that satellite data can go beyond a simple scaling factor of a priori emissions and can be used to constrain mechanisms of soil NO_x emissions. Richter et al. (2005; Figure 1) showed how these observations could be used to diagnose trends in NO_x emissions over the globe. However, there are significant uncertainties in the satellite retrievals and interpretation

which still have to be resolved. The discussion emphasized that these and other results are just beginning to tap into the incredible potential of the satellite data sets.

Processing

NO_x sinks. The hydrolysis of N_2O_5 on aerosols is a significant sink for NO_x , converting it into HNO_3 . In models, approximately 30% of all HNO_3 production occurs through N_2O_5 hydrolysis, with $\text{NO}_2 + \text{OH}$ accounting for 40% and the remainder involving reactions of organic species and NO_x (Liao and Seinfeld, 2005). Uptake coefficients for the hydrolysis of γ on aerosols were originally derived for the stratosphere and used cold sulfuric acid as the aerosol. In the troposphere, aerosol composition is different and both temperatures and relative humidities are higher. Recently, laboratory studies have been undertaken for a better description of the rate of hydrolysis of N_2O_5 within the troposphere. At the workshop, Matt Evans described how these improvements have had a significant impact on the global concentrations of NO_x , O_3 and OH , increasing them globally within a model by 7 %, 4 % and 8 % respectively.

Contributions from Paul Griffiths showed that uptake of N_2O_5 onto mixed phase aerosols containing both organic and inorganic components was slowed by the presence of the organics. This phenomenon has not been considered in global models. Recent advances in observational capability for NO_3 and N_2O_5 have allowed the impact of N_2O_5 hydrolysis to be tested directly in the field and Brown et al. (2006) show values of γ (uptake coefficient) varying by orders of magnitude on ambient aerosol.

Role of NO_3 as an oxidant. NO_3 can act as a potent oxidant during the night time, initiating the oxidation of VOCs. Recent evidence described by Stuart Penkett and Steve Ball from aircraft-borne NO_3 instrumentation and remote DOAS and LIDAR techniques and has shown a ‘cloud’ of elevated NO_3 at the top of the continental boundary layer which was unobservable with previous techniques. The existence of such NO_3 ‘clouds’ may have a significant impact on the processing of pollutants within the boundary layer and on their export to the wider atmosphere.

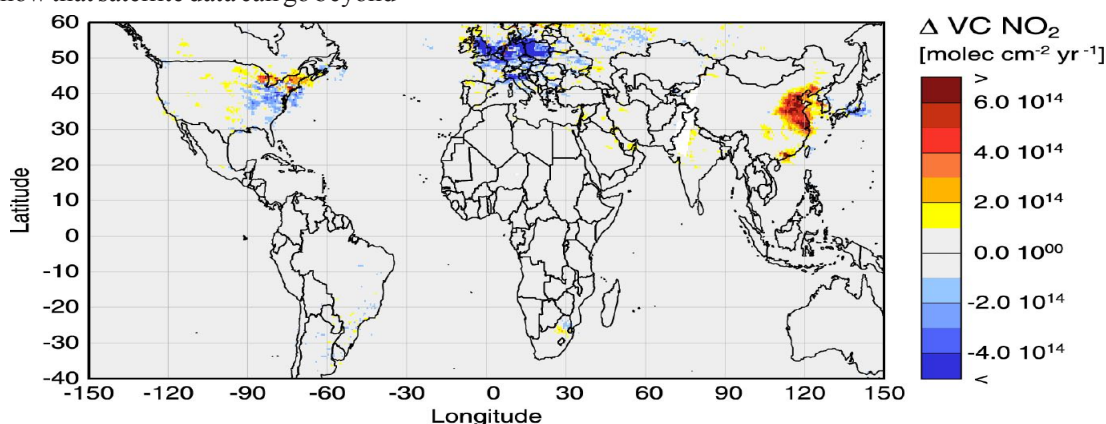


Figure 1. Annual changes in the tropospheric NO_2 column as derived from GOME measurements between 1996 and 2002. Courtesy of Andreas Richter (Richter et al., 2005).

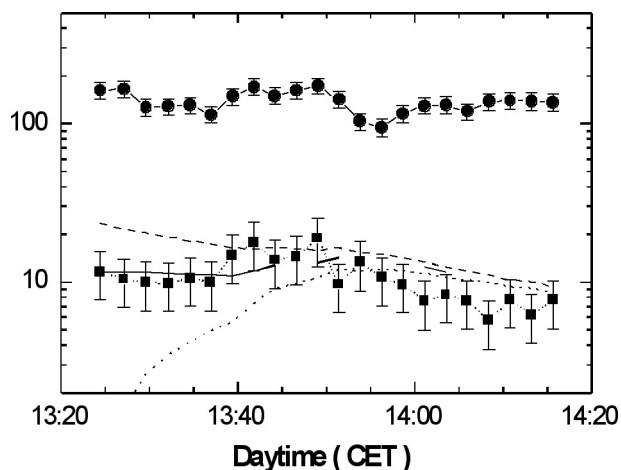


Figure 2. Comparison of measured HONO concentrations with model predictions for a pure gas phase mechanism: —●— denotes the measured HONO data; —■— denotes the photostationary state (PSS) calculation; the different lines represent time-dependent model runs with different starting values: $[\text{HONO}]_{\text{initial}}=0$ (.....), $[\text{HONO}]_{\text{initial}}=[\text{HONO}]_{\text{PSS}}$ (—), and $[\text{HONO}]_{\text{initial}}=2 \times [\text{HONO}]_{\text{PSS}}$ (-----). Error bars on the PSS values denote the accuracy due to experimental uncertainties of the input parameters. Courtesy of Andreas Hofzumahaus (Cox et al., 2006).

Production of HONO. Andreas Hofzumahaus described fluxes of HONO through a forest which were extremely high and difficult to reconcile with current understanding, as it suggests a photo-enhanced heterogeneous source (Figure 2). This source had significant impacts on the HO_x and NO_x budgets within the forest. It is thought that this might involve complex uptake and processing of NO_x on water surfaces within the forested environment.

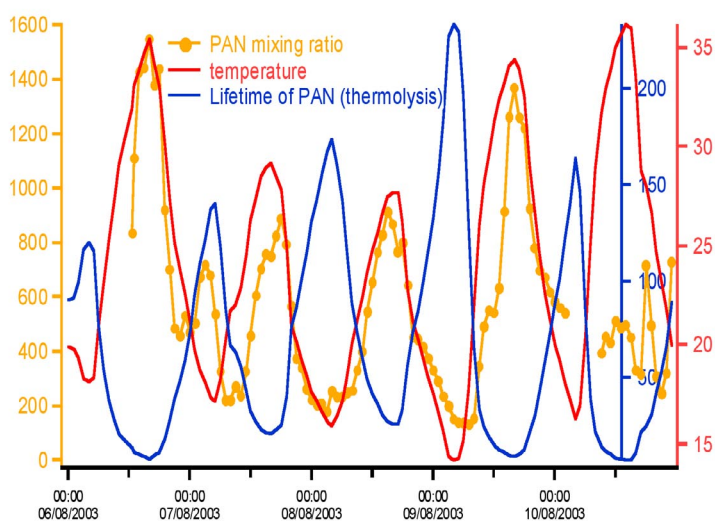


Figure 3. PAN, isoprene mixing ratios and air temperature, and the calculated instantaneous lifetime of PAN over the heatwave period (6th – 10th August, 2003) in the UK (Lee et al., 2006).

Other observations of HONO -- notably from the Polar Regions -- also appear inconsistent with our current understanding of HONO photochemistry (e.g. Beine et al., 2002).

Organic Chemistry. Matt Evans described how observations of high CH_3CHO concentrations in remote areas appear to be inconsistent with observations of PAN and our current understanding of the photochemistry associated with these species. This discrepancy may reflect the difficulties of making observations of CH_3CHO . However, if the observations are correct this poses a severe challenge to our understanding of NO_x and NO_z processing within the atmosphere. The oxidation of complex hydrocarbons (notably isoprene in NO_x rich regions) leads to the production of PANs and other nitrates. The chemistry of these species is complex and poorly quantified. Ron Cohen and Paul Monks (see for example Figure 3) described the impact of this chemistry on the composition of the boundary layer. Mike Jenkin highlighted the uncertainty in the chemistry of these species. The fate (decomposition or deposition) of these species can have a significant impact on the composition of the atmosphere.

Halogen chemistry. Observations of reactive halogen species (notably BrO and IO) would suggest a sink of NO_x through the hydrolysis of BrNO_3 and INO_3 on aerosol surfaces. This sink has been observed in Arctic ozone depletion events. However, quantifying the global significance of this sink is not currently possible owing to a lack of observations of halogen species outside of the boundary layer.

Response of the biosphere to changing emissions. The biosphere will respond to changes in N emissions in a variety of ways and through a variety of mechanisms. For example increased global emissions of N may increase N deposition, enhancing biosphere growth; however it may lead to higher O_3 concentrations which may lead to plant damage inhibiting growth. It is unclear how the biosphere has and will respond to changing nitrogen emissions.

Transport

Convection. Convection is important for determining the global distribution of many species. It causes the vertical redistribution of species, wet deposition of species and the production of lightning NO_x . Global models treat these processes as being to some extent separate and unconnected. Thus, they have a poor ability to accurately represent the extent of vertical transport.

Subgrid issues. A wide range of sub-grid issues exist in determining the composition of the atmosphere. The non-linear nature of the chemistry, in-homogeneity of emissions and depositions, scales of atmospheric motion, etc., all lead to errors in models due to the assumed grid scale. There is currently no systematic way of addressing these issues. Steve Ball described

the impact of small-scale variability on chemical composition, highlighting the impact of isoprene emissions on the small scale distribution of O_3 and NO_3 (see Figure 4).

Boundary layer issues. Our understanding of the movement of material within the boundary layer under stable, low diffusivity situations appears weak with impacts on the surface concentration of species and their distribution through the atmosphere.

Sinks

Ammonia. Ammonia sinks and sources are complicated, highly coupled and variable on very small scales. Thus, the concept of separate ammonia emission inventories and deposition mechanisms are inappropriate.

Organic nitrogen species. The processes by which organic nitrogen species are lost through wet and dry deposition are badly quantified and highly uncertain. The wet deposition of PAN to fogs appears to occur but it is not understood how this occurs. Whether isoprene nitrates are deposited is uncertain but is significant for NO_y , NO_x and O_3 distributions over isoprene rich regions. Uncertainty over these depositional sinks of organic NO_y lead to uncertainty over the global distribution of NO_y and NO_x .

Deposition within models. Deposition schemes within global models are old and do not include recent advances. Stevenson highlighted the substantial difference between models in their attribution of NO_y deposition to wet or dry mechanisms. Uncertainties remain over which processes limit the dry uptake of HNO_3 , and the appropriate treatment of the deposition of aerosols.

Impacts

Anthropogenic nitrogen in the biosphere. The response of the biosphere to increased nitrogen levels is complex. Understanding the full response will require complex coupled models. The answer also depends upon the timescale, the region being investigated and the nature of the problem.

Impact on SOA. Mike Jenkin discussed the impact of nitrogen upon Secondary Organic Aerosol composition and Gordon McFiggins discussed the role of nitrogen upon inorganic and organic aerosols. Both of these influence the ability of aerosol to act as cloud condensation nuclei

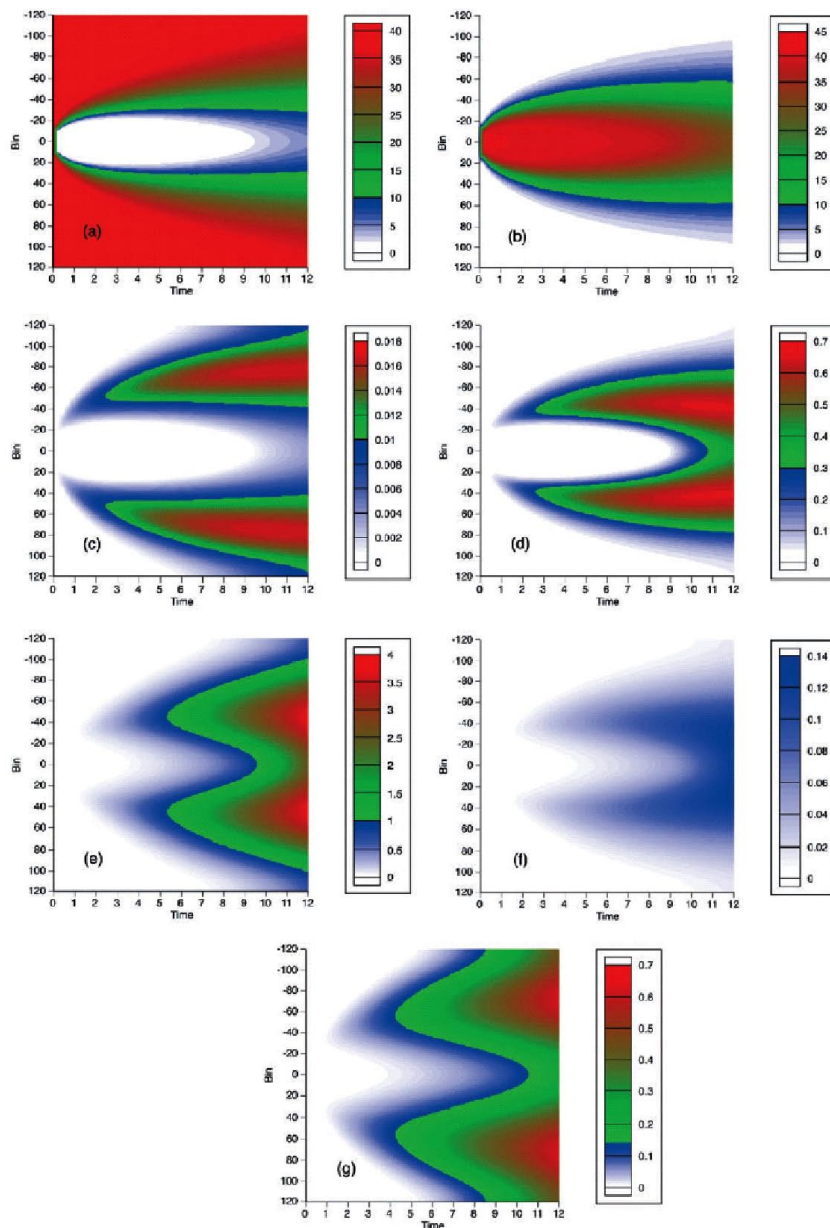


Figure 4. Spatial distributions from the Gaussian plume model of Jones *et al.* (2005): (a) O_3 , (b) NO_2 , (c) NO_3 , (d) N_2O_5 , (e) HNO_3 , (f) products from the reaction of NO_3 with co-emitted anthropogenic hydrocarbons and (g) products from the reaction of NO_3 with biogenic hydrocarbons in the background atmosphere. Note that (f) and (g) are plotted on the same scale. (Courtesy of Steve Ball; Cox *et al.*, 2006)

and the subsequent uptake of water onto these aerosols to form cloud. This has a potential impact upon the aerosol indirect effects and hence climate. These impacts are not considered in current climate models.

What are the impacts of nitrogen species on human health? Lise Frohn discussed work quantifying the impact of nitrate aerosol on human health through the use of linked atmospheric-health models and highlighted to potential of links for the health community.

Conclusions on Global Atmospheric Cycling

Emissions

NO_x emissions in low NO_x environments. More observations of nitrogen species are required in a wider variety of environments to test our current understanding of the processes controlling the emissions of nitrogen.

Satellite observations. More observations of NO_x and NO_y, and in particular their vertical profiles, are required in a wider variety of environments in order to assess satellite retrieval algorithms and to aid in the interpretation of satellite data.

Processing

NO_x sinks. There remain significant uncertainties in the rates of reactive uptake of N₂O₅ onto aerosols. Further laboratory studies of uptake onto different aerosol types under different temperature and relative humidity regimes are needed to reduce this uncertainty. Field observations of N₂O₅ and NO₃ concentrations are needed to test model predictions.

Role of NO₃ as an oxidant. More observations are needed of the distribution of NO₃ over a variety of regions and timescales to assess the extent of the impact of NO₃ chemistry and its vertical distribution.

Production of HONO. Increased observation datasets of HONO within a variety of environments and conditions are needed, as are laboratory studies of the potential heterogeneous production mechanisms for HONO.

Organic chemistry - 1. An assessment of the quality of CH₃CHO observations should be made to validate the observations made in remote regions.

Organic chemistry - 2. The chemistry of the complex organics species produced by the oxidation of isoprene in the presence of NO_x should be studied in more detail both in the laboratory and in the field.

Halogen chemistry. Observations of reactive halogen species over a variety of environments and conditions would allow an evaluation of the impact of halogen chemistry on NO_x concentrations.

Response of biosphere to changing emissions of oxidized and reduced nitrogen. Coupled biosphere-chemistry-climate simulations are needed to assess the impact of changing emissions on the planet.

Transport

Convection. Links between the meteorologists and the atmospheric composition modelling community should be strengthened so that the parameterization of these processes can be done on the best possible footing.

Subgrid issues. Approaches to investigating sub-grid scale issues should be developed, evaluated and considered for implementation into global models of the atmospheric cycling of N.

Boundary layer issues. We should improve our understanding of the transport of species through the boundary layer under stable conditions through field and computer experiments.

Sinks

Ammonia. The emission and deposition of ammonia should be considered as a single process with appropriate parameterization within models.

Loss of organic nitrogen species. More detailed process studies of the removal of organic NO_y from the atmosphere are needed to quantify this issue.

Representation of deposition within models. Recent advances in our knowledge of the fundamental processes leading to the deposition of nitrogen species should be included in models. Mechanisms to represent the sub-grid nature of deposition should also be developed and implemented.

Impacts

The impact of anthropogenic N on the biosphere. Coupled biosphere and atmosphere models are needed to fully understand the impact of anthropogenically emitted nitrogen on the atmosphere. They need to contain a full representation of the chemical and biological systems that process nitrogen.

The impact of nitrogen species on the climate properties of aerosols. The impact of inorganic and organic nitrogen species on cloud properties should be investigated and suitable parameterizations developed for climate models.

The impacts of nitrogen species on human health. The links between the global observational and modelling community and the health impact communities should be strengthened so that each can benefit from the experience of the other. (See accompanying article by Frank Raes).

Uncertainties in the Atmospheric Chemical Processing of Fixed Nitrogen

The principal questions addressed were as follows.

- What new measurements (either in the laboratory or in the field) are needed to test or confirm our understanding of the chemical changes occurring during the atmospheric processing of oxidised forms of nitrogen?
- Which are the key areas where improved chemical parameters and mechanisms are required for our quantitative understanding of the atmospheric processing of nitrogen?
- What are the main limitations in representing the current knowledge of the atmospheric processing of oxidised nitrogen species in atmospheric models?

New measurements required. Much of the discussion of what new measurements are required was related to the problem of understanding the complex chemistry

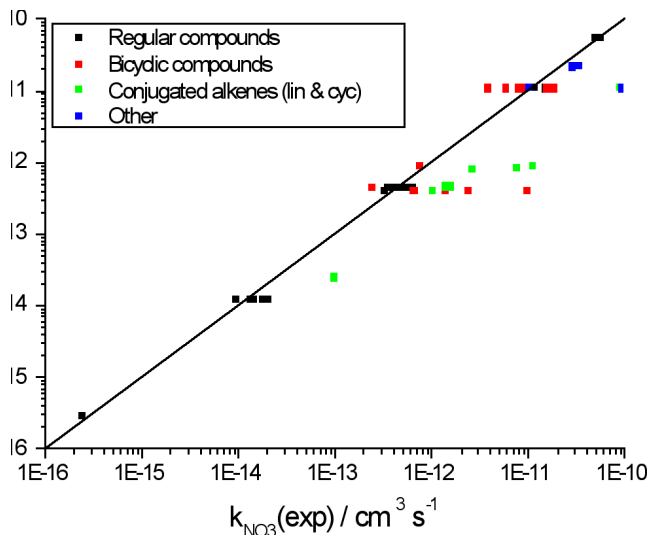


Figure 5. Comparison between the experimental rate coefficients for NO_3 +alkene reactions and rate coefficients predicted using the SAR. “Other” indicates nearly-conjugated alkadienes, where the two double bonds are separated only by a single $-\text{CH}_2-$ group. *Courtesy of Luc Vereecken (Cox et al., 2006).*

occurring not only in the gas-phase but also on surfaces. Implicit with a discussion of desired new measurements is the identification of key areas where an enhanced understanding of the chemical processes involved in the atmospheric transformation of oxidised nitrogen-containing species would lead to the greatest improvements in our quantitative understanding of the atmospheric nitrogen cycle. Specific examples will be expanded upon in the next section.

In terms of gas-phase chemistry, the detailed kinetics of the reaction of OH with NO_2 , to form HNO_3 , are still relatively uncertain (ca. $\pm 30\%$) (Atkinson et al., 2004) and under polluted urban conditions may in fact be the major source of uncertainty in quantitatively understanding the fate of atmospheric nitrogen. In comparison to this well-studied “daytime” sink for atmospheric nitrogen (obviously, HNO_3 can readily be lost from the atmosphere) the nighttime formation of HNO_3 is much less certain – this uncertainty is essentially due to the hydrolysis of the N_2O_5 on aerosol surfaces (Evans and Jacob, 2005). Data were presented which showed that, in laboratory experiments, the efficiency of this process is significantly reduced when oligomeric hydrocarbon material is present in/on the aerosol – presumably with a surface excess. It is becoming increasingly clear that such large molecular weight organic material is intimately involved with the formation and growth of secondary organic aerosol (SOA) material. The kinetics and mechanisms of SOA formation are very uncertain and so, necessarily, are the effects of the formation of oligomeric oxidised hydrocarbon material on the efficiency of N_2O_5 hydrolysis processes. Also related to this area is the potential significant inclusion of (semi- and non-volatile) organic nitrogen-containing species into SOA. Whilst it is clear that the presence of oxynitro-substituents ($-\text{ONO}_2$) in organic species lowers

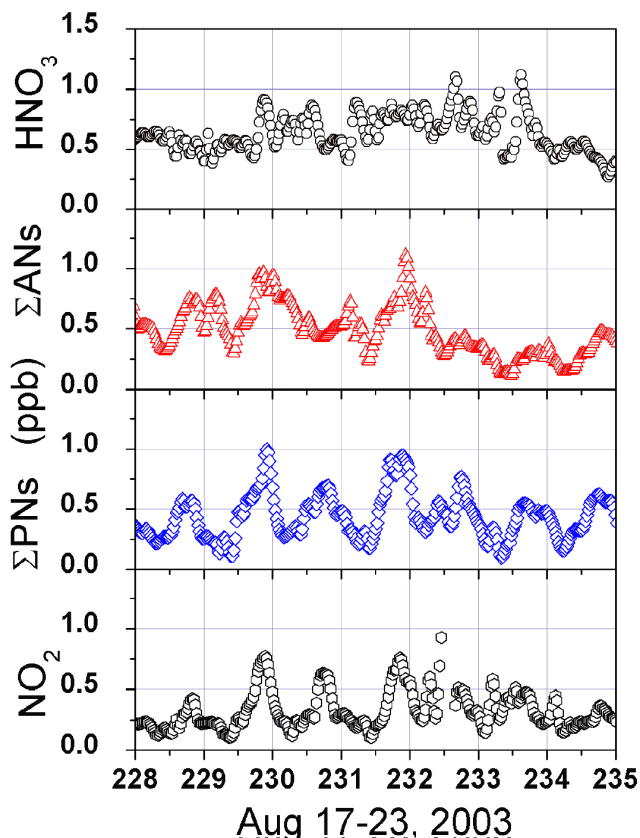


Figure 6a. Example field data for ambient measurements of NO_2 , HNO_3 , alkyl nitrates (AN) and peroxyacyl nitrates (AN) (Cox et al., 2006).

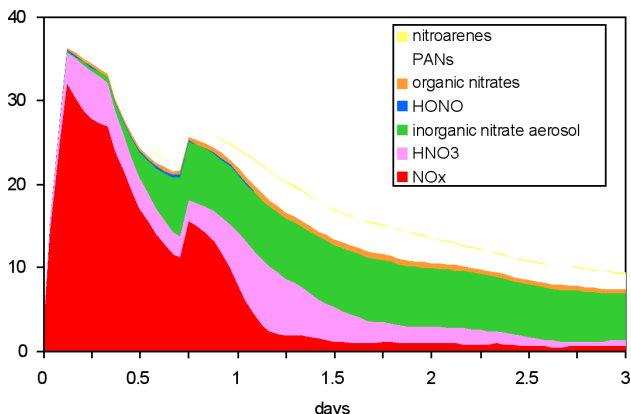


Figure 6b. MCM Simulation of the chemical cycling of oxidised forms of nitrogen as a function of time for an idealised air trajectory. After two days processing, 30% of the components are organic in nature (Cox et al., 2006).

their saturation vapour pressure (and hence increases their propensity to transfer from the gas-phase to a condensed organic-phase), very few such data are available which can be used to formulate reliable estimation methods for these physical properties.

In general, the atmospheric chemistry (formation and degradation kinetics and mechanisms) of organic NO_y species are not well understood. For instance, very few

formation yields of organic nitrate species (RONO_2) from the reactions of organic peroxy radicals (RO_2) with NO , have been measured in the laboratory, yet for atmospheric chemistry it is important to understand how these yields vary with the structure of RO_2 and in parameter-space (temperature and pressure). Those reactions studied also need to be understood in terms of chemical theory in order to be able to reliably estimate RONO_2 formation efficiencies for radical intermediates (very many of which are atmospherically important) which have not and cannot be measured in the laboratory. There was a common theme in the discussion of uncertainties in the atmospheric chemical processing of nitrogen compounds that structure activity relationships (SARs) are ultimately needed in order to be able to predict rate parameters, and ideally product yields, for reactions which cannot be measured in the laboratory. Such SARs are founded on extensive, systematically varied and reliable databases of laboratory rate data. In addition to estimating unknown rate data, SARs can also help to point out where there may be details lacking in the understanding of reaction mechanisms. For example, where a SAR starts to break down – but where it works well for a range of model chemical species – this may be due to secondary effects beginning to govern the kinetics. These may include a change in mechanism – such as evidence for H-abstraction in the reactions of certain alkene species with NO_3 (Figure 5).

Where the understanding of the atmospheric degradation of organic NO_y species (e.g. the reactions of NO_3 with alkenes and the reactions of OH with RONO_2) would certainly benefit from more rate data, they would at least equally benefit from product studies.

During the plenary presentation for the global atmospheric cycle for fixed nitrogen (made by Ron Cohen) field data were presented which represented measurements of the concentration of HNO_3 , the total concentration of alkyl nitrates ($\sum \text{RONO}_2$) and the total concentration of peroxyacyl nitrates ($\sum \text{RO}_2\text{NO}_2$) (Figure 6a). These measurements were made by thermally dissociating ambient air samples (over prescribed temperature ranges) and detecting the concentration of liberated NO_2 by laser induced fluorescence (LIF). Such measurements afford an unprecedented opportunity for comparing measured ambient organic NO_y data with the results of simulations using very detailed chemical mechanisms, for example the Master Chemical Mechanism (MCM) (Figure 6b).

Key Areas for Research

Areas where directed research would most benefit our quantitative understanding of the atmospheric chemical processing of nitrogen compounds concerned much of the material mentioned in the previous section in addition to others where a picture of the chemistry is just emerging. In terms of the established view of atmospheric nitrogen cycling, laboratory kinetic and mechanistic data concerning the atmospheric formation and processing of organic NO_y would be extremely beneficial, particularly in understanding the efficiency with which nitrogen is cycled between NO_x and NO_y forms. The links between semi-/non-volatile organic NO_y species and SOA need to

be elucidated – for example, measurements revealing the significant presence or absence of multifunctional organic nitrate species in ambient fine aerosol material would be extremely illuminating.

In terms of emerging areas of research, the global atmospheric scope of surface-mediated, photosensitised formation of HONO from NO_2 needs to be established. In a more indirect, but by no means insignificant, sense the reactive involvement of NO_x in alkene ozonolysis reactions needs to be elucidated. For example, it is now well established that this class of reaction can be a major – and indeed sometimes dominant – source of HO_x (Heard et al, 2004) radicals but it is not at all known if the carbonyl oxide (Criegee) intermediates from which, for example, OH is formed can react with NO (Ariya et al 2000, Paulson and Orlando, 1996). Criegee intermediates are similar in structure to organic peroxy radicals and hence are expected to react with NO (yielding NO_2 and a carbonyl). If these bimolecular reactions are sufficiently fast then, under given atmospheric conditions, they may effectively out-compete the decomposition of Criegee intermediates – i.e. these latter reactions may not be a significant direct source of HO_x radicals in the atmosphere. The NO_x -dependence of OH yields from ozone-alkene reactions need to be investigated (Calvert et al., 2000). The experimental complication associated with such studies – and the reason that none have been reported to date – is that ozone (O_3 ; i.e. one of the reactants) is reactive towards NO . Related to this area of chemistry is the NO_x -dependence of SOA yields from the ozonolysis of monoterpene species, for example α -pinene. Data presented by Neil Donahue (Donahue et al., 2005) at the Faraday Discussion meeting that showed the SOA yield (expressed on a mass basis) from the reaction of ozone with α -pinene decreases significantly in the presence of added NO . It is not clear whether this latter effect is due to reactions of α -pinene Criegee intermediates with NO (giving rise to more fragmentation and the production of more volatile reaction products in the degradation mechanism) or due to a perturbation of the complex peroxy radical-peroxy radical reactions which occur in the absence of NO in the majority laboratory experiments (i.e. NO converts peroxy radicals to alkoxy radicals).

Representing Nitrogen Processing in Atmospheric Models

The inclusion of the detailed processes described above in atmospheric models was discussed but a significant point is that the size of chemical modules within global models is not likely to increase significantly in the near future. This is due, in part, to the need to significantly increase/enhance other modules within models, such as those describing aerosol chemistry and physics. Again, on the theme of global modelling, although the overall aim is to represent complex chemistry efficiently, the chemistry of species such as isoprene needs to be represented more explicitly. Obviously, this requires a better experimental and theoretical understanding than we currently have of the atmospheric oxidation of this very important hydrocarbon and its interactions with the atmospheric nitrogen cycle. In essence, to most reliably produce

efficient, reduced chemical mechanisms it is necessary to start with very detailed, “trusted” mechanisms against which smaller mechanisms can be tested.

Conclusions on the Atmospheric Chemical Processing of Fixed Nitrogen

Much is already understood about the atmospheric processing of oxidised nitrogen species. However, understanding of the following chemical processes is particularly uncertain:

- The role of developing SOA material in changing the rate of N_2O_5 on aerosol surfaces
- The detailed chemistry of organic NO_y species – both their formation and degradation
- The significance of multifunctional organic nitrate species for the formation and growth of SOA material
- The atmospheric significance of HONO formation from surface-mediated, photosensitised reactions of NO_2
- The perturbation of direct HO_x formation and SOA formation from the ozonolysis of alkene-species

In order to reduce some of these uncertainties, proposed research activities include:

- Further laboratory studies of the kinetics (rate measurements) and mechanisms (product measurements) of the reactions of various RO_2 with NO ; the reactions of NO_3 with various alkenes and the reactions of OH with various $RONO_2$ species. The temperature- and pressure-dependence of such data would also be of benefit.
- The systematic measurement of saturation vapour pressures of a variety of organic nitrogen-containing species (particularly alkyl and peroxyacyl nitrates) would be of benefit in formulating structure property relationships.
- Additional studies of the conversion of NO_2 to HONO on surfaces would be very instructive.
- New experimental designs are needed to study the radical and molecular products of the reactions of ozone with various alkenes in the presence of NO_x .
- Finally, detailed chemical mechanisms such as the Master Chemical Mechanism should continue to develop and evolve with the state of knowledge of the constituent chemistry, and mechanisms should be tested against the best field data available. Additionally, large atmospheric models should be encouraged to incorporate the most up-to-date chemical schemes.

Surface – atmosphere exchange of fixed nitrogen.

This topic is discussed separately in the accompanying

paper by David Fowler and colleagues (Fowler *et al.*, 2006).

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The Nitrogen Cycle and Its Influence on the European Greenhouse Gas Balance

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Background

The nitrogen cycle is of fundamental importance in human health issues, ecosystem functioning and global change: it provides a key control of the global carbon cycle through effects on primary production and decomposition; it is a major determinant of terrestrial and aquatic biodiversity; it affects particle and other chemical production in the atmosphere; and it has major impacts on greenhouse gas fluxes and stratospheric ozone depletion. It is therefore

a matter of great concern that global cycling of reactive nitrogen (NH₃, N₂O, NO₃⁻), N_r, is estimated to have more than doubled (Vitousek et al., 1997; Galloway et al., 2004), whereas, by comparison, the C cycle is less than 10% perturbed by human activities (IPCC, 2001). Despite this concern, much less effort has been given recently to quantifying the nitrogen cycle than to the carbon cycle. This may be partly due to the apparent simplicity of the “carbon story” and the dominant role of CO₂ as a greenhouse gas. By contrast, the complexity of multiple interactions and impacts makes the nitrogen problem less accessible to a wide audience. This has not been helped by previous research efforts on N_r being widely dispersed between the different N_r forms and their impacts: e.g., research on N₂O, NO and NH₃ fluxes have been considered separately, as have studies on N_r in atmospheric chemistry and N_r impacts on ecosystem functioning. It is therefore a major scientific challenge to bring together these issues and provide a clear picture of the role of nitrogen in global change.

This has already been recognized within the Biosphere Atmosphere Exchange of Pollutants (BIAFLUX) Joint Research Programme in the frame of the ACCENT¹ Network of Excellence. In this context, an integrated research project has been established to address the core aspects of reactive nitrogen in the atmosphere. This project, NitroEurope IP², or NEU in short, is funded by the European Commission under the 6th Framework Programme and brings together more than 60 institutions all across Europe and beyond.

The Nitrogen Challenge

There is a clear need to translate process understanding into quantitative models that can address interactions with other global change drivers, that can be applied in relation to practical land management decisions and that can be up-scaled to the whole of Europe to support the development of European sector policies. It is important to note that greenhouse gases provide the main focus of the NitroEurope project, but N_r interactions with biodiversity, atmospheric chemistry and water must be recognized where they occur. In this respect, the approach provides a first step towards an eventual multi-effect assessment related to nitrogen.

It is crucial to keep a balance between addressing the prime issues of European N budgets in relation to C cycling and greenhouse gas exchange, while at the same time being aware of the interactions with other environmental issues. A key point of integration is the recognition that climate change policy requires integrated assessment of net greenhouse gas exchange (NGE) rather than just CO₂. This is vital for future strategy development, since approaches that maximise CO₂ uptake may not optimize NGE (Li et al., 2005). Apart from the obvious links between N and C cycles, there is a requirement to assess overall ecosystem nitrogen budgets, since other N_r losses, e.g. NH₃ emissions and leaching of nitrate (NO₃⁻), are considered as indirect sources of N₂O emissions

¹ <http://www.accent-network.org/>

² <http://www.nitroeuropa.eu> for more information on the project

under the IPCC methodology (IPCC, 1996). The need to integrate the analysis of N_r and GHG at linked field-, farm- and landscape-scales is thus recognised, including the consideration of spatial interactions with NH_3 emissions and NO_3^- leaching. Furthermore, N_r gases can form aerosols which affect the radiation balance of the earth. Aerosols are not a focus of this call. However, the contribution of aerosol biosphere-atmosphere exchange to N deposition and aerosol production/loss within canopies is needed to calculate N_r budgets and NGE. Similarly, interactions with ecosystem functioning and biodiversity must be considered in order to understand the observed responses of NGE to global change drivers.

This new approach will advance the state of the art substantially across a wide range of “Nitrogen Science”. Key examples are given here in relation to: quantification of biosphere-atmosphere exchange, novel approaches to reducing key uncertainties in GHG emissions, flux measurement techniques, responses to global change, process modelling at plot scales, landscape analysis, regional upscaling and the verification and uncertainty of regional estimates.

Networks for measurement of biosphere-atmosphere exchange

Individual components of the deposition budget of atmospheric N_r are currently derived within the UNECE/EMEP framework, the US National Acid Deposition Programme (NADP) and the US Clean Air Status and Trends Network (CASTNet), for example from wet deposition and through indirect inference from air concentration measurements. However, in contrast to CO_2 , no networks currently exist for direct measurements of N_r fluxes, e.g. with micrometeorological approaches. A project within the European LIFE programme demonstrated the feasibility of long-term (dry) deposition monitoring of the different N flux components at three sites (Erisman et al., 2001), and several EU projects have recently addressed the exchange of a sub-set of atmospheric N compounds within individual ecosystem types, such as reduced N with grasslands (GRAMINAE; Sutton et al., 2001) and oxidized N (incl. N_2O) with forests (NOFRETETE³).

Similarly, annual exchange fluxes of non- CO_2 GHGs have mainly been measured within a number of unrelated national activities. For example, the recent EU GREENGRASS⁴ project represented one of the first trans-European studies of NGE, focusing on managed grasslands. A major advance over past and present measurements will be achieved through this project by establishing the first network for long-term measurements of N_r pools and fluxes and NGE, which is comprehensive, collocated, highly integrated and covers each of the main European ecosystems and climatic zones. As part of the analysis of the data from this network, the development of inferential approaches to derive dry deposition from air concentration measurements is taken forward, by adding compensation point approaches and through calibration

³ <http://195.127.136.75/nofretete>

⁴ <http://www2.clermont.inra.fr/greengrass/>

and verification over a wide range of (European) ecosystems.

Techniques for the measurement of trace gas and aerosol fluxes

The long-term flux determination of trace gases requires continuous measurement techniques for the surface/atmosphere exchange of the different N_r species and GHGs. The most direct micrometeorological technique, eddy-covariance (EC), is routinely applied to measure CO_2 exchange. However, EC requires high-precision, fast-response sensors, which, for most N_r species, are in their infancy. For example, the first ever eddy-covariance measurements have only recently been presented for NH_3 (Famulari et al., 2005) and particulate N (Nemitz et al., 2004). Because of these limitations, fluxes of N_r species are usually measured with gradient approaches, which rely on empirical parameterizations and similarity assumptions. While cost-efficient gradient monitors are available for NO and NO_2 , current automated systems for artefact-free determination of gradients of gaseous NH_3 are labour-intensive to operate and have only recently begun to be extended to measure HNO_3 and aerosol NO_3^- and NH_4^+ . A new generation of photo-acoustic NH_3 monitors (e.g. Pushkarsky et al., 2003) are a promising reliable alternative to wet-chemistry based systems, but a cost-effective solution optimized for ambient flux measurements is still lacking.

Fluxes of CH_4 and N_2O are routinely measured with manual or automatic chamber techniques, which have the advantage of easily resolving concentration changes. By contrast, these average over only small areas (e.g. $<1 m^2$) and modify the microclimate of the enclosed area. Recent developments have now demonstrated that, in high emission areas, long-term measurements of N_2O and CH_4 can be made by eddy-covariance using tunable diode laser absorption spectroscopy (TDL-AS) (Scanlon

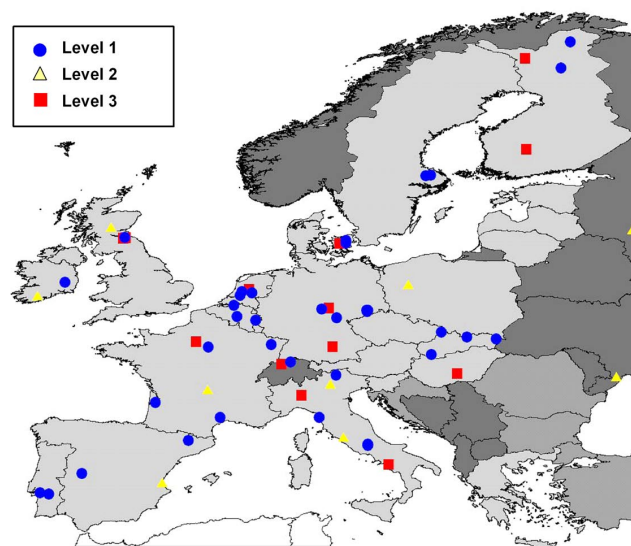


Figure 1. Overview of the measurement network of the NitroEurope IP (EU light grey, candidate countries medium grey, non-EU countries dark grey)

& Kiely, 2003; Di Marco et al., 2005). Principal European research groups are involved in method and instrument development for N_f fluxes and are integrated to further develop state-of-the-art flux measurement techniques.

Key uncertainties in Net Greenhouse Gas Exchange (NGE)

While CO_2 exchange is measured in fairly extensive global networks (CarboEurope IP⁵, FLUXNET⁶), the effect of N inputs on carbon sequestration remains poorly quantified. Nevertheless, increased terrestrial C sinks due to increases in N availability are likely to have contributed to the increase in forest growth rates observed across Europe which account for some of the 'missing global carbon sink' (e.g. Hunter and Schuck, 2002). Initiatives to model the global effect of N availability on C sequestration (e.g. Holland et al., 1997; Asner et al., 2001) are limited by uncertainties in the mechanistic understanding of C-N interactions in plant and soil and a lack of field data for verification. Although a range of N fertilisation experiments have been performed at the plot level, site networks that derive both CO_2 net ecosystem exchange (NEE) and N uptake are extremely limited (Nadelhoffer et al., 1999). An initial comparison of measurements of NEE with rough estimates of N deposition at the Ameriflux and Euroflux sites indicates that C uptake peaks at a N input of about 20 kg N ha⁻¹ yr⁻¹ (Holland et al., 2004). Robust estimates of N deposition need to be added through flux measurements to existing CO_2 measurements at 22 sites and infer deposition at another 50, which will allow a rigorous statistical assessment of the effect of N deposition on C sequestration across a site network.

Soils are the main source of N_2O (55-65%) and CH_4 (60-70%) in the atmosphere, with nitrifiers, denitrifiers and methanogens considered the most important microbial sources (Conrad, 1996). The relative importance of nitrification, denitrification and nitrifier denitrification in producing or consuming NO and N_2O is a key to understanding the mechanism of trace gas N emissions from soils and for accurately up-scaling and quantifying the N_2O source strength of the EU terrestrial biosphere, which is still poorly constrained. Current simulation models like DNDC, Animo and Century are able to 'explain' at best 50% of the measured daily variation in N_2O fluxes over time. A better understanding of the basics of N_2O production will contribute to improving our plot scale models and estimates of the EU greenhouse gas budget, and subsequently to devising strategies to minimize emissions. The same applies also to CH_4 , where the competition between the anaerobic process of methanogenesis and the oxidative process of CH_4 -oxidation controls the net exchange at the soil-atmosphere interface. Based on recent developments, novel enrichment methods are applied and further developed, making use of single- and double-¹⁵N-labelled NH_4NO_3 and ¹⁸O-labelled water (H_2O) to differentiate between N_2O production from nitrification, nitrifier denitrification

and denitrification, and to further quantify emissions pathways such as plant-mediated N_2O losses via the transpiration pathway.

Responses of N_f and GHG fluxes to global change

The exchange of greenhouse gases and reactive nitrogen between the biosphere and the atmosphere is largely controlled by external drivers such as climate, deposition, management and land use and will therefore be significantly affected by the predicted future changes in these drivers. Carbon dynamics and GHG exchange are affected by changes in N availability, as it is a key element which determines the functioning of many ecosystems (Vitousek, 1997). Consequently, increased atmospheric N deposition and CO_2 content, changes in climatic drivers such as temperature and precipitation patterns, occurrence of extreme events, changes in the water table and changes in agricultural management practices such as use of fertilizers, and changes in land use such as afforestation and land abandonment will all affect C-N interactions. Well documented features include changes in soil N status (e.g. Gundersen et al., 1998; Emmett et al., 2004; Ritter et al., 2003; Goodale and Aber, 2001) and C sequestration (e.g. Jackson et al., 2002; Cao and Woodward 1998; Lindroth et al., 1998; Christensen, 1999), with consequent changes in nutrient cycling characteristics (e.g. Jussy et al., 2002) and GHG exchange (Priemé et al., 1997, Vitousek et al., 1997; Christensen, 1999; Ambus and Robertson, 1999). The impacts of these driver changes across European ecosystems are poorly understood and have been hampered by a lack of integrated system-level studies. Default emission factors have been used as the best available practice (e.g. IPCC 1996). However, there is increasing evidence that this methodology is often not correct (e.g. Skiba et al., 1996; Jungkunst et al., 2004; Fiedler et al., 2005).

The effect of such changes will be studied through the employment of field-scale manipulations across the European continent in order to assess their geographical importance. Previous studies on responses to global change have tended to separate by ecosystem, land use, driver and climatic region, leading to different approaches by the agricultural and semi-natural ecosystem science communities. These communities are brought together through the Nitro Europe IP project, encouraging debate and improving comparability. Furthermore, the project adds measurements of N budgets, CH_4 and N_2O to a significant number of existing major experiments which investigate the effect of system changes on CO_2 exchange. In particular, included is a new generation of experiments that look at the combined effect of various drivers, such as CO_2 , temperature and water. The importance of such multi-factorial experiments in contributing to our understanding of ecosystem responses to climate change is becoming increasingly apparent (Shaw et al., 2002). Through this highly cost-effective integration it will be possible for the first time to compare the effects of multiple drivers and their interactions on NGE Europe-wide.

⁵ <http://www.carboeurope.org/>

⁶ <http://www-eosdis.ornl.gov/FLUXNET/>

Process modelling of C and N fluxes at the plot scale

The exchange of C- and N-trace gases between soils and the atmosphere is the result of simultaneously occurring production and consumption processes (Conrad, 1996), with C and N cycles closely linked. Recent publications show that increased C sequestration in agricultural soils due to changes in field management may enhance N₂O emissions (Six et al., 2004; Li et al., 2005). Therefore, an integrated research and modelling approach is needed to address the complex interactions between the C and N cycles.

Production and consumption of C- and N- trace gases in soils are predominantly due to the microbial processes of mineralization, nitrification, denitrification, methanogenesis and CH₄-oxidation. Some of these processes are strictly aerobic or oxidative, whereas others are facultative or obligate anaerobic processes. The predominance of a given anaerobic or aerobic process is regulated by soil diffusivity and thus also by soil moisture and other soil properties such as texture and soil organic carbon (SOC) content. Furthermore, management measures are affecting soil microbial processes and thus GHG exchange, e.g. through drainage, irrigation or fertilization. This already complex pattern is further controlled by other biotic and abiotic processes (e.g. Papen and Butterbach-Bahl, 1999; Kaiser et al., 1998), which change substantially on spatial as well as on temporal scales (e.g. Davidson et al., 1998).

In view of the complexity of the involved processes, regional and global scale estimates of the source strength of soils for even the primary greenhouse gases N₂O and CH₄ remain highly uncertain. This uncertainty may be significantly reduced by the further development and testing of process-oriented models at a plot level. In the recent past several biogeochemical models such as the DNDC (Li et al., 1992) have been developed, which are able to mimic the complexity of processes observed in field and laboratory studies and to simulate not only GHG exchange but also other processes, e.g. nitrate leaching or NH₃ volatilization. In several recent EU projects such as the NOFRETETE and GREENGRASS, it has been demonstrated that these models are very useful tools for validating our knowledge of processes. They are already capable of simulating a wide variety of ecosystems, but further development is still needed for improving the predictive capabilities of these models, e.g. with

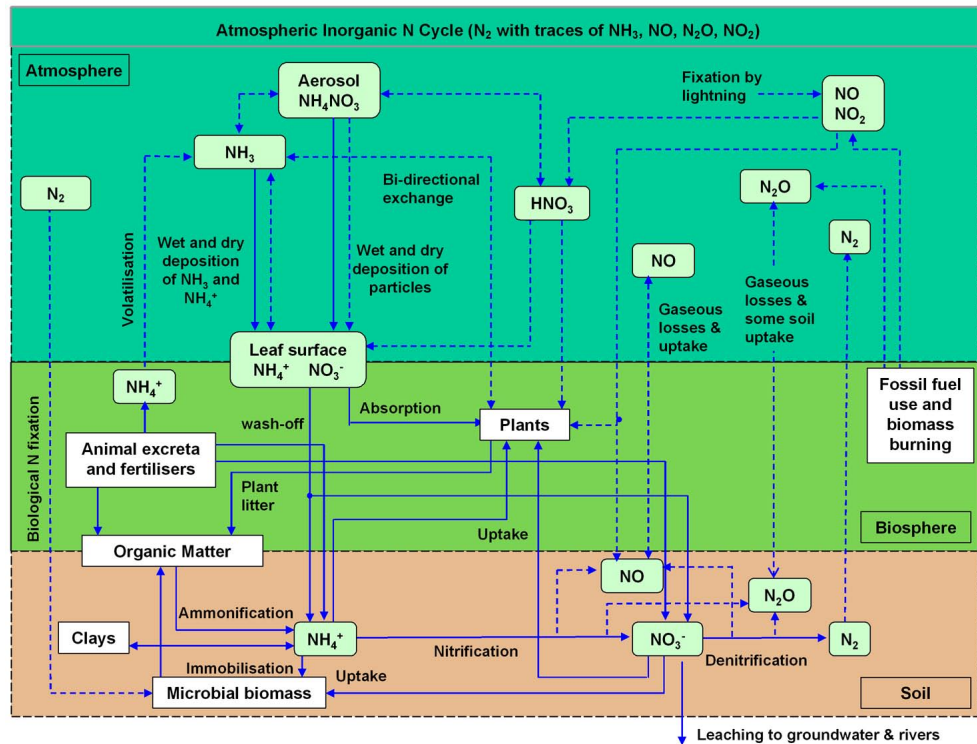


Figure 2. Schematic of the N cycle, distinguishing between gaseous fluxes (dashed lines) and non-gaseous fluxes (solid lines).

regard to the simulation of management activities and their impacts on C and N cycling or with regard to the numerical description of microbial and plant physiological processes. For the first time standardized schemes for assessing model uncertainties will be established and the outcomes of the uncertainty analyses will be used to guide further developments, to ensure that models not only gain in complexity but also in predictive accuracy. Model development and testing will be facilitated by bringing together advanced multi-ecosystem models such as DNDC and several sectoral models. The models will be used for testing hypotheses and to support and guide experimental work. Furthermore, they are a major tool for retrospective assessments of global change impacts on GHG exchange for selected sites across Europe for which mitigation strategies will also be developed.

Landscape and farm level analysis of N_r and GHG fluxes

Most studies on GHG have either addressed fluxes at plot scales (e.g. micrometeorological studies) or at the regional scale, where it is assumed that regional estimates can be made by a simple summation of component emissions from different field sources. Both these scales miss the interactions due to lateral fluxes between source and sink areas in the landscape (e.g. Loubet et al. 2001). Until now, little attention has been given to quantifying N_r and GHG fluxes at the landscape scale. The landscape is defined here as the scale at which adjacent fields, farms and forests etc. interact spatially, typically over distances of <50 m to 5 km, due to changes in surface or ground conditions, or to N transfer by farm management.

Significant advances in the direction of landscape-level assessment have been taken by hydrological researchers, who necessarily consider all the N inputs and fluxes at a catchment scale. However, many catchment studies focus on a larger scale (e.g. several 100 km²) and most apply a semi-distributed modelling approach whereby sources in sub-catchments are simply summed rather than explicitly treating the lateral fluxes between adjacent sources and sinks (e.g. INCA model, Wade et al., 2002; SWAT model, Grizzetti et al., 2003). Only relatively recently have the finer-scale interactions been considered using fully-distributed hydrological models (Ewen et al. 2000; Lowrance et al., 2000; Beaujouan et al., 2002). Of critical importance, however, for the landscape scale is the recognition that the dispersion processes occur both in the atmosphere and in water, pointing to the need for linked atmospheric dispersion and hydrological modelling. Similarly, while the dispersion processes may operate between sources and sinks (e.g. from a livestock farm to neighbouring semi-natural areas, Dragosits et al., 2002), the decisions concerning farm management, which affects both the magnitude and location of emission activities, have to be taken in the context of wider business units. As a result, landscape analysis needs to incorporate a whole-farm perspective.

The inter-disciplinary approach required for landscape-scale analysis represents a major challenge. The first landscape models for integrated analysis of N_r fluxes (NH₃, N₂O, NO₃⁻) have recently been reported (Bernsten et al., 2003; Theobald et al., 2005). These studies combined plot-scale ecosystem models, farm models and dispersion models for agricultural areas, including atmospheric dispersion as given by Theobald et al. and hydrological dispersion as given by Bernsten et al. This will be further advanced under the Nitro-Europe IP Project by bringing together the leading teams to develop the first complete, spatially explicit landscape model for N_r and GHG. This development will draw on the component ecosystem models that will be developed and improved and validated against flux measurements. The anticipated model will incorporate interactions between C and N for both agricultural and semi-natural ecosystems, nesting farm building sources and decision making, and linking to atmospheric and hydrological dispersion.

In parallel, the first Europe-wide network of landscapes for C and N assessment will be established. Landscape inventories will allow the comparison of land-use structure and socio-economic differences, while providing necessary inputs for the landscape model application. The landscape model will be validated through spatial field measurements, which will quantify lateral fluxes and identify unexpected emission “hotspots”. These measurements will advance our knowledge of C-N interactions substantially, as these focus on integrated measurements (air, plant/soil and water). Finally, the modelling and measurement will together provide the basis for considering land management scenarios that utilize “landscape buffering” (Ryszkowski et al. 2002; Sutton et al., 2004) and farm-scale integration as a basis to explore N_r and GHG mitigation potential.

European upscaling of N_r and GHG exchange

There are presently several estimates of European-scale land use related emissions of NH₃, NO_x, N₂O and CH₄, and of the N budget, focusing on nitrate leaching and runoff to surface waters (Bouwman et al., 1997, 2002; Stohl et al., 1996; Freibauer, 2003; van Drecht et al., 2003; Mosier et al., 1998). However, these estimates are all based on a coarse approximation of the needed inputs (e.g. country statistics or data at 0.5 by 0.5 degree resolution; van Drecht et al., 2003) and IPCC-like emission factor approaches (Bouwman et al., 1997, 2002; Mosier et al., 1998) or simple empirical models (Freibauer, 2003). Much of the information required for the derivation of N_r and GHG emissions from agriculture is not included in current European databases. In addition, there are coarse scale land-use change reconstructions available for Europe for the period 1900–2000 (Ramankutty and Foley, 1999). There are currently ecosystem models available that provide process-level descriptions that can be applied to derive spatial N and GHG fluxes at regional scale (e.g. Butterbach-Bahl et al., 2004), but the up-scaling is based on crude assumptions regarding model inputs and results are not yet validated. Also, the different sources of error have not been specified, which is crucial to improve and understand the results and possible biases. As a result, studies on the responses of N_r and GHG emissions to European-scale land-use changes are still lacking.

To overcome this, datasets on NH₃, N₂O and CH₄ measurements obtained within the NEU project and from the literature will be used in combination with detailed ecosystem models for daily NH₃, N₂O, CH₄ and CO₂ emissions (Li, 2000; Li et al., 2000), to derive simplified process-based and empirical models in an integrated multi-sector, multi-component model. Both detailed and simplified bottom-up process-based modelling approaches will be applied to use an integrated approach to estimate the past, present and future N_r and GHG emissions and sinks in response to various scenarios reflecting: (i) past and present land use changes and land management decisions and (ii) various policies and actions that affect N_r emissions in interaction with GHG emissions. These estimates will be based on a linkage of: (i) detailed GIS-based assessment of environmental data (land use, soil type, average climatic situation, altitude, etc.) and farming data (farm types and agricultural management) and (ii) detailed reconstructions of land use changes in the period 1970-2000 and projections for the period 2000-2030 with both detailed process-based models and the integrated multi-sector multi-component framework. To exploit the full potential of available farming data, information obtained from networks, national surveys and farm scale questionnaires will be used to downscale the regional statistics using appropriate disaggregation techniques. Together with these advances, the uncertainties in estimates of European emissions of N and GHG as produced by the bottom-up GIS-based results will be quantified, including verification against independent measurements and a comparison with results from inverse modelling.

European verification and uncertainty

There is a need to make an inventory of available independent measurements and models that can be used for verification, to develop upscaling procedures that produce comparable numbers, and to assess their uncertainty. Furthermore, formalized statistical procedures need to be developed which are suited to analyse the absolute magnitude and supposed trends in official values compared with the independent estimates, in relation to agreed quantitative acceptability criteria. A verified framework for targeting policies (relevant for the IPCC/UN/FCCC and UNECE) is clearly needed to reduce the negative environmental impacts of N and GHG. This will focus primarily on the GHG emissions, while making the connections for eventual full integrated assessment of the full range of nitrogen effects.

During the last few years the first attempts have been made to derive independent top-down GHG emission estimates for individual European countries by inverse modelling (Manning et al., 2003, Vermeulen et al., 1999, Bergamaschi et al., 2004a). The use of inverse modelling for verification of national bottom-up inventories has recently been reviewed and discussed at an EU workshop (Bergamaschi et al., 2004b). While these first studies clearly demonstrate the feasibility of using inverse modelling for verification, a major limitation of these estimates has been the very limited number of continental regional measurements with high temporal resolution. Furthermore, it has been recognized that it is mandatory to use several independent models, in order to get reliable estimates (Gurney et al., 2002). The inverse modelling approach will be extended to Europe, using a larger set of European observations. A European network of tall towers is now established within the EU project CHIOTTO⁷, which will provide quasi-continuous atmospheric monitoring data of greenhouse gases (including CH₄ and N₂O). With heights of 100 to 200 m, these integrate the signal from emissions within regional footprints of typically 500 to 1000 km and therefore achieve good European coverage. Based on this, five independent atmospheric models will be developed and applied for N₂O and CH₄, comprising both Eulerian and Lagrangian dispersion models. This will allow a realistic estimate of overall uncertainties in the top-down estimates.

Methods for quantifying and analysing the uncertainty in outputs of environmental models are becoming increasingly rigorous and efficient (e.g. Van Oijen et al., 2005) and will be applied to the models. Probabilistic techniques, including Monte Carlo assessment of uncertainty propagation from inputs to outputs and Bayesian calibration will be employed. The uncertainties of the estimates of European emissions of N and GHG, produced by the bottom-up GIS models, will be analysed and attributed to the underlying uncertainties in model parameters and input scenarios. The extent to which the uncertainty in the estimates decreases when running the models on high spatial-resolution data for specific regions will be quantified. Uncertainties regarding modelling approach will be addressed by comparing

the results of the bottom-up GIS models with emission estimates produced by top-down inverse modelling and by calculations based on independent measurements. The different emission estimates will be combined into one overall estimate of European N and GHG emissions, with quantified uncertainty.

Independent verification is a vital issue for the EU and its member states in the context of the Kyoto Protocol for GHGs, as well as the Gothenburg Protocol for NO_x and NH₃. In particular, independent approaches are needed for the verification of official national and EU inventories, as this is the only means to assess compliance with agreed emission targets. At present, this remains a topic in its infancy requiring both independent estimates and development of verification procedures that are both scientifically and legally robust. The necessary independent estimates will be provided, including results of bottom-up GIS models, top-down inverse models and comparison with independent datasets. At the same time, procedures for uncertainty analysis will be interfaced with the legal requirements of verification, in order to derive an improved basis for compliance assessment under the international protocols. Finally, the results of the verification assessment and uncertainty analysis, together with the direct data from the experimental studies will be used to update the default methodologies recommended by the IPCC (1996) and the UNECE Task Force Emission Inventories and Projection (TFEIP) (EMEP/CORINAIR, 2004) to calculate national N_r and GHG emissions.

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⁷ <http://www.chiotto.org>

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Joint Research Programme on Aerosols: Air Quality & Climate

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Abstract

The ACCENT Joint Research Program on “Aerosols: Air Quality & Climate” aims to promote research and networking activities in the areas of air quality and climate with specific focus on the role of aerosols, or particulate matter. A series of programs have been put in place to advance our understanding of a number of key aspects in these two thematic areas and to identify the key areas of overlap between air quality and climate studies from a research and policy perspective.

Introduction

Two issues provide the impetus for much of the current research in the field of atmospheric aerosols: (1) aerosol-cloud-climate issues; and (2) the relationship between particulate matter, poor air quality, and adverse human health effects. While these issues may seem somewhat removed from one another, there are areas of significant overlap. Climate issues imply a global scale, whereas human health issues are often thought of as local phenomena. Health and climate issues have also generally been dealt with by different scientific communities. Issues of scale and scientific discipline have, to a large extent, served to maintain the separation between these topics, to the detriment of both. In reality, there is a good deal of common ground between these issues and there is an urgent need to develop coordinated research activities in a number of areas common to climate, air quality and health.

The activities within this Joint Research Programme are centered on addressing the challenges of developing

coordinated research strategies and programs for the air quality, health and climate communities, linking the local, regional and global scales. More specifically, the program aims to:

- determine specific issues related to aerosols and clouds that are common to the topics of climate, air quality and human health;
- determine the most pressing measurement and modelling needs in these areas and develop specific programs to address them;
- devise joint measurement and modelling strategies that will produce information that is necessary and useful in all three areas;
- link parallel activities within ACCENT to create a forum for information exchange between the air quality, health and climate communities, with aerosols and clouds as a focus;
- compile and make available a summary of current nationally-funded research programs, within Europe, in the areas of aerosols and clouds. Investigators interested in climate, air quality and health issues can make themselves aware of work going on in countries other than their own, and can facilitate contact between researchers in Europe.

Research Workshops & Activities

To date, and from a research perspective, a number of workshops have been held within ACCENT on Constituents of Organic Aerosols; Formation, Dynamics and Growth of Atmospheric Aerosols; and Effect of Chemical Composition on Warm Cloud Droplet Activation. The meeting on Constituents of Organic Aerosols was a one-time meeting to summarize the current state-of-the-art in characterization of organic aerosol properties and resulted in the production of a workshop report (Dye and Myhre, 2005). The Dynamics and Growth of Atmospheric Aerosols activities comprised a series of workshops covering a 3 year period with the aim to produce clear research outcomes with respect to developing and quantifying our understanding of the new particle formation process. Similarly, the Effect of Chemical Composition on Warm Cloud Droplet Activation activity also comprises a series of workshops, again covering a 3-year period.

The general goals of the Dynamics and Growth of Atmospheric Aerosols activity are to develop models that are adequate for use in global climate models starting from molecular level understanding and to develop suitable instrumental tools and observation strategies to quantify atmospheric new particle formation (Petäjä and Kulmala., 2006). The main foci areas are modeling of aerosol formation, computational chemistry, ion-induced nucleation, nucleation experiments and theory, nuclear molecular clusters, nucleation involving organics and sulphuric acid, and composition of freshly nucleated particles. Some recent experimental advances include reduction of the lower detection for neutral clusters from

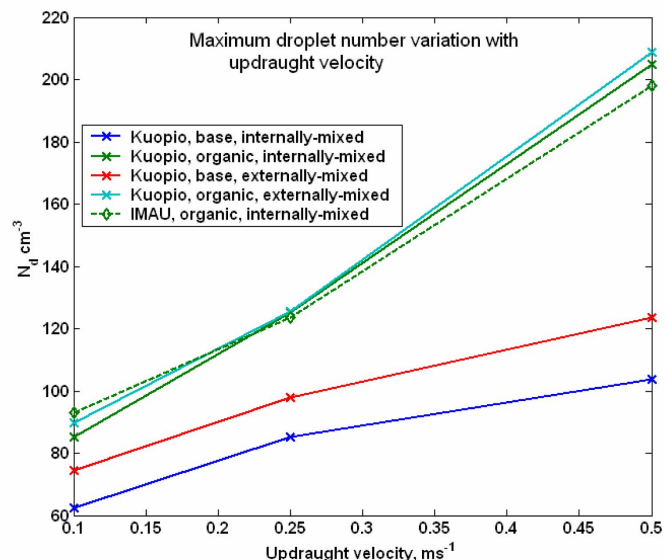


Figure 1. Predicted activated number of cloud droplets as a function of updraft velocity for two of the models (University of Kuopio & IMAU, University of Utrecht). Base case is for inorganic marine aerosol externally and internally-mixed and the organic case is organic matter added either internally or externally to the base case aerosol. The scenarios are those summarized in O'Dowd et al., 2004, for North Atlantic marine aerosol.

3 nm to 2 nm (Kulmala, manuscript in preparation), an analysis of the global importance of atmospheric nucleation (Spracklen et al. 2006 ACPD), and global observations of the existence of ion clusters (Kulmala et al., manuscript in preparation).

The motivation behind the Effect of Chemical Composition on Warm Cloud Droplet Activation activity series was to quantify the key tools and processes required to address the effect of chemical composition on cloud droplet activation in warm clouds and to evaluate these in terms of identifying the most appropriate way forward for developing aerosol-cloud parameterizations for inclusion in large scale models. There exists many flavors of aerosol-cloud-chemistry (0-D) process models, particularly in terms of the treatment of thermodynamics: such models include, for example, Pitzer molality-based ion interaction treatment extended to include small organic ions; equilibrium constant models (EQUISOLV II) extended to include an ideal treatment of organic components; formulations using osmotic coefficients varying with composition; and extended versions of ADDEM¹ which treats multicomponent inorganic/organic aerosol activation using the Pitzer-Simonsen-Clegg mole fraction model, combined with UNIFAC² modified to be applicable to atmospherically representative compounds. In addition, some of the models have explicit detailed treatment of mass and heat fluxes.

¹ ADDEM - Aerosol Diameter Dependent Equilibrium Model

² UNIFAC - UNiversal quasichemical Functional group Activity Coefficients

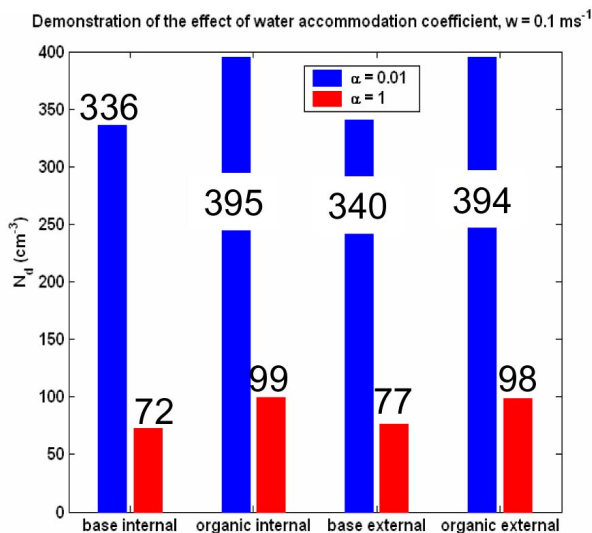


Figure 2. Difference in predicted activated number of cloud droplets for an accommodation coefficient choice of 1 and 0.01.

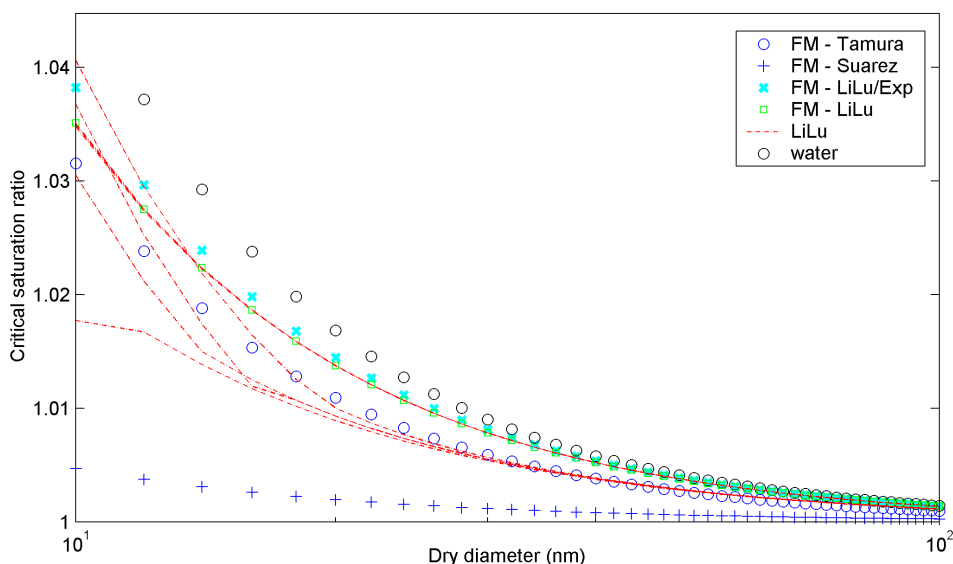


Figure 3. Critical saturation ratio versus dry diameter for the ‘multi 4’ composition using a variety of surface tension models. ‘FM’ – multi-component organic surface tensions calculated using the Fainermann and Miller mixing rule. ‘Tamura’, and ‘Suarez’ and ‘LiLu’ – use of the Tamura mixing rule and Suarez thermodynamic model to represent the binary organic surface tension. ‘Binary/Szykowski’ – use of the binary LiLu model to represent the binary organic tension of all organic components except Suwannee River Fulvic acid where the Szykowski equation is employed. ‘water’ – using the surface tension of pure water; ‘FM/LiLu’ – use of the Fainermann and Miller mixing rule and LiLu binary model (dash lines - representing a range of permutations of the coupled inorganic/organic LiLu model.)

The models are undergoing evaluation to determine the most appropriate thermodynamic module to deal with cloud droplet formation from inorganic-organic mixed aerosols. In addition, different treatments of surface active material inclusion are also being evaluated. By inclusion of a fully characterized sub-micron organic aerosol fraction, the suite of models is exploring warm cloud activation of multicomponent aerosols.

The initial studies have focused on marine aerosol physical and chemical properties (O’Dowd et al., 2004) and later studies will focus on continental organic-inorganic aerosol mixes as well as organic-inorganic aerosol mixes found under very polluted conditions influenced by mega-cities.

Significant findings have emerged from the first inter-comparisons between the cloud parcel models, where it has been demonstrated that there is significant sensitivity of predicted droplet number to aerosol composition, less sensitivity to mixing-state assumption and thermodynamic treatment of the components, and the greatest sensitivity to kinetics of droplet growth as represented by the accommodation coefficient of water.

The most challenging, complex and computer-intensive component of these process models is of course the thermodynamic module, and the selection of the most appropriate module can be a significant topic of debate. However, the impact of thermodynamic module is far out-weighted by the apparently simple choice of mass accommodation coefficient for water – an issue also of long standing debate.

Figure 1 displays the number of droplets activated on the marine aerosol population typical of the North East Atlantic. The base case corresponds to a base case comprising sea-salt and nss-sulphate aerosol internally and externally mixed. Note that this is the typical mixture that has been generally assumed for sea salt in models, but as shown by O’Dowd et al. (2004) and Quinn et al. (2005) a significant fraction of marine aerosol mass is organic. Thus the base case here is compared to one where organic aerosol mass has been added (see O’Dowd et al., 2004 for detailed discussion). The predicted activated droplet concentration is seen to be significantly increased by the addition of organic mass whether internally or externally mixed. However, also shown is that the predicted concentration by two very different models from the University of Kuopio and IMAU, irrespective of the state-of-mixing, demonstrate only small differences in the resulting droplet concentration.

The models were re-run with an accommodation coefficient³ of 0.01 (as opposed to 1.0 in the base case) and it was found that that this parameter has the

³ The accommodation coefficient can be viewed as the probability of a molecule sticking to a surface if it collides with that surface. In the current discussion, this use of the term refers to the accommodation coefficient of water vapor.

most significant impact on the predicted activated concentration (Figure 2). While the issue of the most appropriate value of accommodation coefficient has been an issue of discussion for decades, there clearly seems to be two camps preferring different choices. In response to this difference of opinion, the Aerosols Joint Research Programme is holding a workshop on *Accommodation Coefficient: Current Consensus* in April 2007.

A number of techniques for predicting the surface tension of multicomponent aerosol have also been developed. These have been systematically tested against laboratory measurements of binary and multicomponent aqueous solutions of inorganic / organic mixtures. As may be expected, all of the systems studied showed a reduction in surface tension with increasing solute concentration, and this was qualitatively reproduced by all predictive techniques. The quantitative understanding of mixed organic and mixed inorganic/organic systems has been shown to be limited, however. Predictive techniques only have the necessary skill to accurately capture multicomponent surface tension if binary solution data are available. Of the purely predictive techniques, a coupled thermodynamic approach to surface tension prediction was generally the most satisfactory, but this was composition dependent. Obviously, further laboratory studies of representative binary and higher order mixtures in conjunction with ambient measurements of chemical and physical properties of aerosol particles are necessary.

Critical saturation ratios as a function of dry size were predicted for all multi-component systems studied (Figure 3). Deviations between predictions increased as the dry size decreased. Similarly, it was evident that using the surface tension of pure water, rather than calculating the influence of the solutes explicitly, lead to consistently higher value of the critical saturation ratio. A neglect of the compositional effects lead to noticeable differences even at large dry sizes. The Figure illustrates such a sensitivity with 4 different model formulations for a particular multicomponent mixture. The FM/LiLi technique was best able to capture the measured surface tension behavior for this mixture (Topping et al., 2006). It should be noted that such predictions ignore the potential reduction in Raoult effect resulting from partitioning of surface active species between the bulk and surface of the activating droplet (Kokkola et al., 2006). Detailed experimental verification of the existence of such an effect is essential to understanding the effect of organic components on droplet activation.

Young Scientists Exchange Programme

In 2005 and 2006 two Exchange Programmes were launched for young scientists with the aim of promoting technology and knowledge transfer in the priority areas of the Joint Research Programme on Aerosols. One such exchange program involved the National University of Galway, Ireland, Max Planck Institute for Meteorology, and the European Commission's Environment Institute at the Joint Research Centre. The aim of this exchange was to implement the aerosol dynamics module M7 (Vignati et al, 2004) in the regional climate model

REMOTE (Langmann et al., 2000) and to elucidate the production of sea-spray aerosol over the north Atlantic through testing of new combined inorganic-organic seas spray source functions. Following implementation and testing of new source functions on the regional scale, the most appropriate source function will be implemented in the global chemical transport model TM5. This work is conducted in close association with the European research project MAP (Marine Aerosol Production, <http://macehead.nuigalway.ie/map>).

Preliminary work in this area has implemented M7 and is extending the module to interface with a combined inorganic-organic source function developed from a combination of in-situ eddy covariance sea-spray flux and detailed chemical composition measurements. In this approach, the organic aerosol component of sea-spray (Yoon et al., 2006) is related to satellite derived chlorophyll-a concentration at the ocean surface. Figure 4 illustrates the first simulations of total (organic-inorganic) sea-spray mass in the accumulation mode over the oceanic regions around Europe for the month of June 2003.

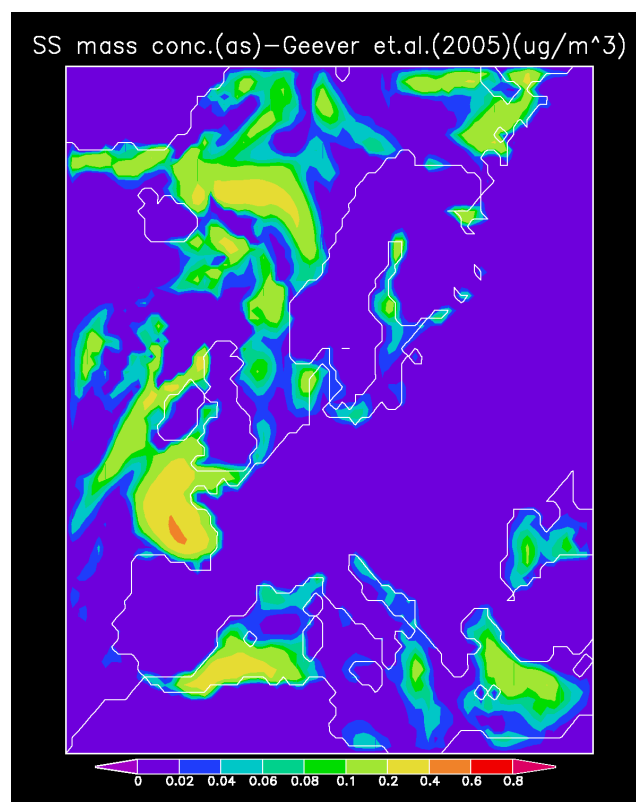


Figure 4. Combined inorganic-organic sea-spray accumulation mode mass distributions for selected month of June 2003. The source function is derived from a combination of eddy-covariance flux measurements (Geever et al., 2005), chemical composition measurements (Yoon et al., 2006) and chlorophyll-a concentrations derived from satellite.

Strategic Activities

One of the most significant meetings to be held under the Joint Research Programme on Aerosols was a meeting to define the Common Issues between Air Quality and Climate Change (in terms of aerosols), from both a research and policy perspective. The outcome was a Position Statement on the current state-of-the-art and Recommendation Report on future directions (Hansson and O'Dowd, 2006).

Background: Research, monitoring, and policy development in the areas of Air Quality and Climate Change have largely developed as separated processes. This has occurred for a variety of reasons including the differing life-times and spatial scales associated with the main pollutants and the perception of impacts. However, the most recent studies highlight that Air Quality and Climate Change have many overlapping and inter-linked problems, or common issues, suggesting that Air Quality and Climate Change should be handled in a coupled manner from the research and the policy perspective. It is thus of common interest to both scientific and policy communities to be aware of these common issues. The Joint Research Programme on "Aerosols: Air Quality & Climate" had identified four key questions on Common Issues in research and policy aspects of aerosol impacts on Air Quality and Climate Change. These were considered at a "Common Issues" workshop, held in Dublin, January 2006, where the current Position on Common Issues from the research and policy communities was developed and an assessment of future Common Issue recommendations was produced.

The main conclusions and recommendations are outlined as follows: Integrated Air Quality & Climate Change policy is essential to optimize the environmental effectiveness and investment required in end-of-pipe, structural, and behavioral change measures. Consideration of the Common Issues will result in significant economic savings in terms of abatement and mitigation strategies.

Climate sensitivity, i.e. the projected warming due to doubling of GHG concentrations, is a key concern in the present development of climate policy. However, direct and indirect aerosol radiative forcing also contribute to climate change, so uncertainties in these forcings propagate to uncertainties in global radiative forcing, significantly influencing how accurately climate sensitivity can be determined. Climate Change can also impact on Air Quality and exacerbate the impacts of air pollution on human health, agricultural production and ecosystems. Such connections imply the need for increased integration in the understanding of air quality and climate change common issues in order to develop the appropriate synergistic abatement strategies.

The recent establishment of the hemispheric task force on long-range pollution transport and its impacts on air quality provides an incentive to address air quality issues in a similar domain as climate change. This development provides a basis for further integration of these issues and exploration of win-win scenarios in relation to abatement options for both areas.

Recommendation 1. From the process level, there still lies considerable scope for the improvement of emission inventories (including aerosol formation) adjusted for use at different scales. Significant research effort in this area, and integration of research with emissions data from statutory regulation, is strongly recommended to improve input and output of the air quality and climate change models. The development of a process by which data obtained for regulatory purposes are combined with emissions data from other sources is required and should respond to the needs of advanced models as well as basic reporting requirements.

Recommendation 2. Aerosol formation, transformation, and removal processes need to be further elucidated. Air quality and climate change models must interface better. Improved urban-to-regional scale integration of models (i.e. nesting) is needed, given that an urban-scale spatial resolution down to 1 km x 1 km is a necessity (at least in locations where urban populations are dense). In addition to improved nesting, improved treatment of aerosol parameters such as mass, number, chemistry, hygroscopic and optical properties is also a requirement.

Recommendation 3. Monitoring strategies for air quality and climate change observation systems should be developed in a harmonized manner to ensure comparable databases of key Common Issues measurements. In particular, the modeling and monitoring of aerosol number concentration and the contribution of combustion aerosols (in particular "soot") to the aerosol population is essential to facilitate better descriptions of the physical and chemical processes of particles, thereby, enabling the possibility of future studies to quantify aerosol impacts on health and climate.

As both climate and air quality are undergoing notable change, and given that the two systems are not decoupled, it is strongly advisable to develop research and policy in these two systems in an integrated manner.

Conclusions and future activities

A combination of one-off workshops, workshop series driving coordinated research, young scientists exchange programs and strategic research-policy interaction meetings have been conducted during the first half of the ACCENT Joint Research Programme activity period. These activities have produced new results on directions in the areas of aerosol formation, organic aerosol characterization, aerosol-climate interactions and research-policy synergy development. The second phase will aim to focus more on the urban-to-regional scale interactions and on the impact of air quality on health. A summary of activities and reports can be found on the ACCENT web portal.

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Climate Change and Air Pollution – Research and Policy

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Politics, Policy and Science

The Earth System Science Partnership¹ calls for integrated approaches to understanding the Earth System (including interactions between the environment and society), and it seeks to develop a framework for global stewardship in order to respond to the challenge of global change. As one of the projects of IGBP, the IGAC communities' contributions also should aim to fulfil this larger goal, in the area of atmospheric chemistry.

Over the years the IGAC community has increasingly contributed to Earth System science. However to date it has not been active in the aspect of stewardship that includes policy making, and at the end of the day, political action. We believe this aspect of stewardship should also be actively considered within our work. As a step in this direction, the ACCENT project has included such discussions in its programme. We hope that this will assist other projects, such as IGAC, at also moving in this direction.

In a democracy, citizens are represented by politicians who are elected by the people to take decisions for the common good. The legislative texts which politicians need to decide upon are prepared by policy makers – that is, civil servants in national ministries, or governmental institutions like the European Commission. Such preparation usually includes broad consultation with stakeholders.

Scientific research on issues of immediate relevance to society – such as the environment or human health – undoubtedly plays a role in policy making. Scientific research observes the functioning of the world, unravels relationships between human activities and effects that are not immediately obvious, and hence may discover or even predict problems that require political action. This scientific understanding is often critical for finding solutions to the identified problems. The scientific community, through its link with education, also plays an important role in raising public awareness of the issues.

The tasks of scientists, policy makers and politicians can be linked using the concept of “risk” – defined as the chance of losing capital, including quantifiable and non-quantifiable capital. Scientists identify risks and relate them to human activities (risk assessment), while policy makers develop practical solutions to mitigate or adapt to these risks (risk management). Politicians eventually decide which solutions to adopt, and hence they determine the level of risk to be accepted (risk taking).

¹ ESSP is comprised of the WCRP, IGBP, DIVERSITAS, and IHDP programs. IGAC is one of several projects under IGBP. See <http://www.essp.org> for more information.

The scientific community must defend its methods of producing knowledge tested by critical and reproducible observations. But at the same time they must be aware and open to the notion that people and politicians also use other yardsticks to guide their decisions. For example, the setting of environmental targets is not based solely on rational cause-effect relationships unravelled by scientific research; economic, social and ethical aspects are considered as well. This is often seen as a negotiating process – which it is, but it stems in theory from the desire of politicians to reduce the overall risks to society, not simply on any one issue.

An increasing number of commentators are calling upon the scientific community to be responsive to non-specialist interest groups, and to involve them in developing the applied science agenda necessary to more effectively contribute to policy making. The views of scientists on which issues policy makers and society should be informed about, are generally well represented in the scientific literature. However, we believe that the views of society should also be more clearly presented in the scientific literature, that is, if we are serious that science should be responsive to the needs of society.

The problem in this two way communication may partly lie with editors of scientific journals, but undoubtedly it also lies with policy makers who don't reach out to the scientific community with their questions. The underlying problem may be the lack of a suitable platform for science-policy communication.

Below, we sketch the increasing integration between research and policy in the areas of air pollution and climate change, where Integrated Assessment Models (IAMs) provide a common platform for science-policy discussions. Some such IAMs can be considered operational precursors to the Earth System models advocated by Earth System science. In recent years, several proposals have been made to progress Earth System modelling, from models of intermediate complexity to place-based research. Here we illustrate how it is instructive to consider specific societal problems or policy areas, to see how science, policy and society communicate, and to consider how progress has been made with and through integration.

Climate Change and Air Pollution

The problems of climate change and air pollution have, to a large extent, a common cause: emissions from fossil fuel burning. Up until now however, these two problems have been addressed by separate policies. Here we explain the reasons for this separation but also describe how the natural and socio-economic sciences are intersecting to support more integrated and effective policy.

The basics behind both issues are well established. Society's demands for energy, transport and commodities have been easily satisfied by the availability of cheap fossil fuels. The combustion of fossil fuel leads to emissions of greenhouse gases (GHGs): carbon dioxide in particular, but also methane and nitrous oxide. GHGs accumulate in the atmosphere, warming its lower layers and producing climate change, which in turn impacts the Earth System

as a whole. Carbon dioxide is chemically rather inert and hence difficult to remove from combustion emissions. The main strategy for reducing carbon dioxide emissions therefore – and thus the cornerstone of any long-term climate policy – is emission prevention. Prevention can be achieved by structural changes in the energy sector (i.e. improved efficiency and carbon-free renewable energies) and by behavioural changes (i.e. reduced energy use).

Fossil fuel combustion also emits conventional air pollutants: carbon monoxide, volatile organic compounds, carbonaceous aerosols (“soot”), nitrogen oxides and sulphur dioxide. Some of these compounds react in the atmosphere to form secondary pollutants such as ozone and particulate matter (PM; e.g. sulphate, nitrate and organic matter), which impact ecosystems and human health. Developed countries have reduced these impacts initially by improving energy production efficiency and, more recently, using cheap end-of-pipe emission control technologies.

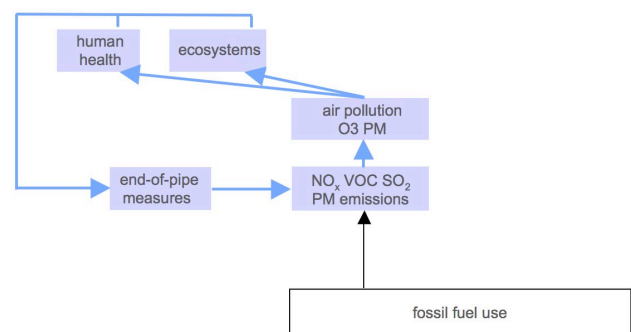


Figure 1. Air pollution policies.

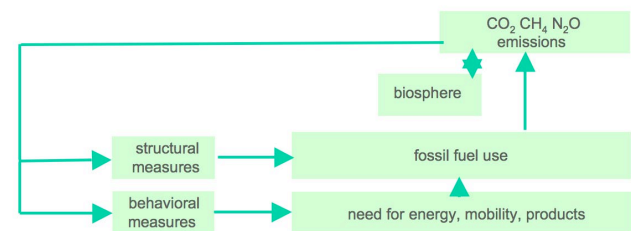


Figure 2. Climate change policy: reducing emissions.

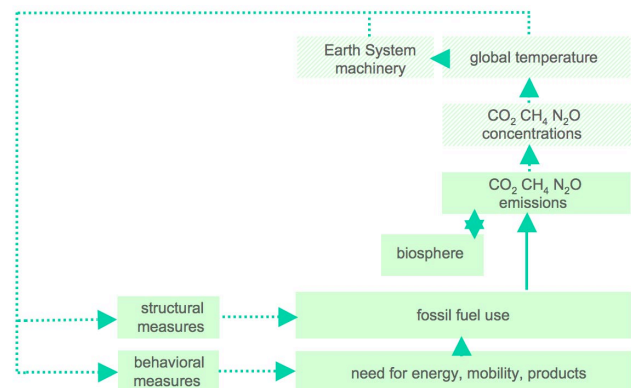


Figure 3. Climate change policy: avoiding dangerous interference.

Current Policy Separation

Reducing fossil fuel use would address both climate change and air pollution. However, fossil fuels provide over 80% of the current global energy supply and will continue to be the dominant energy source for decades to come. Furthermore, GHGs differ from conventional air pollutants in their physico-chemical properties, leading to differences in the relative atmospheric longevity and ease of removal from combustion effluents. Currently and in recent decades, therefore, separate policy frameworks have been developed for air pollution and climate change (see Figures 1,2 and 3).

Air pollution policy has developed faster than climate change policy, primarily because of cheap end-of-pipe control technologies for air pollution derived from fossil fuel combustion (Figure 1). Successful air pollution abatement is also due to the short lifetimes of pollutants (i.e. aerosols and tropospheric ozone), which means that impacts occur relatively close to emission sources, hence allowing local-regional scale solutions. Conversely, long-lived GHGs mix throughout the global atmosphere, with climatic effects that cannot be traced back to specific sources or regions. Climate change policy therefore requires global commitment, irrespective of local-regional emissions and impacts.

Towards Policy Integration

In recent years it has been increasingly recognised that air pollution and climate change are linked in several ways, and that they could be beneficially addressed by integrated policy (Swart *et al.*, 2004). The push for policy integration comes mainly from consideration of implementation costs. This is particularly true in developed countries, where cheap air pollution control technologies are already widely implemented, and further reductions are likely to require structural and behavioural measures. In developing countries current economic growth and the supporting development of energy production systems provides the opportunity to tackle air pollution and GHG emissions simultaneously. Immediate investments in existing carbon-free energy technologies would seem to be the solution. However, for major developing countries it is economically attractive to use large domestic coal reserves, necessitating the use of end-of-pipe technologies both for air pollution and for GHGs. For GHGs, however, end-of-pipe technologies such as carbon dioxide capture and storage are still in the research and development stage.

Policy integration means finding the mix of end-of-pipe, structural and behavioral measures that meet air pollution and climate change targets at the lowest cost. Finding this optimal mix is achieved using integrated assessment models (IAMs), which describe some or all of the interactions shown in the figures. For air pollution policy IAMs consider the blue boxes of Figure 1 and optimise for the cheapest mix of end-of-pipe controls that meet human health and ecosystem condition targets. For current climate change policy (where targets are expressed as GHG emission reductions, cf. Kyoto Protocol) IAMs consider the green boxes in Figure 2, and optimise for the

cheapest mix of structural and behavioural measures that meet emission reduction targets.

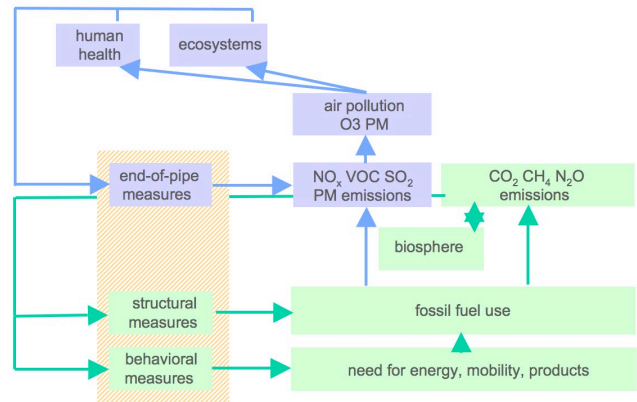


Figure 4. First level integration: optimizing mix of policy measures.

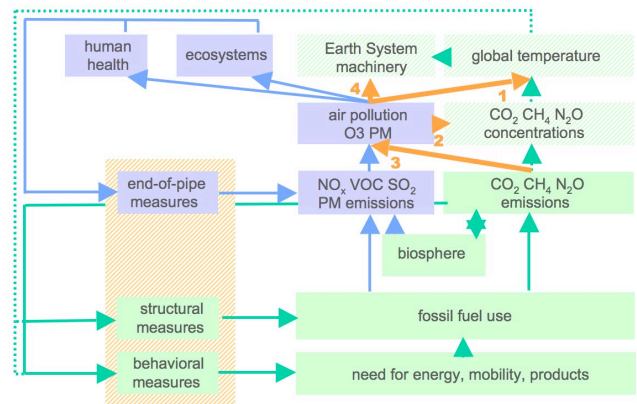


Figure 5. Second level integration: consider also atmospheric/ climate processes.

First-level policy integration requires IAMs to consider the blue and green boxes in Figure 4 simultaneously, optimizing for the cheapest mix of end-of-pipe, structural and behavioural measures that meet both GHG emission and air pollution targets. However, the ultimate goal of climate change policy is not to reduce GHG emissions, but to “avoid dangerous anthropogenic interference with the climate system” (UNFCCC, 1992). Hence climate change policy should consider both the solid green and shaded green boxes in Figure 3. Second-level integration – integration of “dangerous-interference-avoidance” policy with air pollution policy – would require consideration of all boxes in Figure 5, and optimisation for the cheapest mix of end-of-pipe, structural and behavioural measures that avoid dangerous interference with the climate system and that meet air pollution targets.

For second-level integration, several processes in the atmosphere-climate system must be considered – processes by which air pollution can affect climate, and climate change can affect air pollution. These processes (see orange arrows in Figure 5) include:

1. The radiative impact of conventional air pollutants: ozone is a potent GHG, whereas aerosol particles – depending on their chemical composition – either reflect or absorb solar radiation (Houghton *et al.*, 2001);
2. Methane, together with nitrogen oxides and volatile organic compounds, is an important precursor for background tropospheric ozone, hence methane emission reductions performed as a climate policy will also affect tropospheric ozone (Dentener *et al.*, 2005);
3. The reduction of troposphere ozone would reduce the oxidizing capacity of the troposphere; this in turn would enhance the atmospheric lifetime and concentrations of methane (Dentener *et al.*, 2005);
4. Aerosol pollution may affect cloud and precipitation formation, and hence impact local and possibly regional atmospheric circulation and the water cycle (Menon *et al.*, 2002; Andreae *et al.*, 2004).

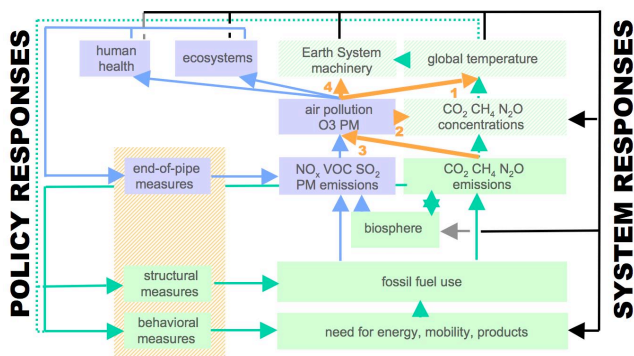


Figure 6. Policies: one more feedback in the Earth System.

Moving from optimizing either air pollution or climate change policy only to first-level and then second-level integration requires increasingly complex IAMs. In Europe, the RAINS model² has been used to optimise air pollution policy. RAINS already integrates economics and technology with atmospheric chemistry, since before the impacts of air pollutants can be calculated, a good description is required of their atmospheric transport, transformation and eventual removal. The POLES model³ – a model of European and global energy demand and supply with a carbon dioxide emission module – has been used to develop climate change policy. Other than the carbon dioxide emission module, POLES contains no description of the climate system. A first-level integration might be possible by combining RAINS and POLES. However, second-level integration would require the inclusion of a climate model able to calculate the effect of GHGs and conventional air pollutants on the climate system. Models with this capability are still

² www.iiasa.ac.at/~rains/home.html

³ web.upmf-grenoble.fr/iepe/Publications/publicRech5.html

under development, but do not lend themselves to the multiple calculations required in optimisations. Models of intermediate complexity (Cloussen *et al.*, 2000) are therefore required.

In the integrated system of Figure 5, policies are a feedback that aim to control the effects of fossil fuel combustion.

There are also inherent feedbacks in such a system (see Figure 6); for example, the effects of pollution or global warming on various components of the system such as the biosphere or society. Considerable research is required to understand just how strong policies can be or should be, given the existence of such inherent feedbacks. IAMs used in policy making to date are unsuitable for such research as they do not capture the full complexity of the system. However, integrated models are emerging that couple economics, atmospheric chemistry, climate and ecosystems, and which through sensitivity runs, are starting to explore the effects of policies and inherent feedbacks (Prinn *et al.*, 2006). It appears therefore, that air pollution and climate change policy and science are now mature enough to allow a systematic approach to policy integration. This provides an example of concrete progress in Earth System modelling.

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Learning about Inter- and Transdisciplinarity in Atmospheric Sciences: Training of Early-Career Scientists in ACCENT

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In order to tackle an environmental problem, different scientific fields (e.g., air pollution modelling, analytical chemistry, toxicology) need to be involved (interdisciplinarity). The collaboration between academic disciplines is, however, not sufficient, as the expertise required is not restricted to the academic world alone, and other parts of society (e.g., politicians, the media) must participate (transdisciplinary collaboration). This paper describes the organisation, outcome and lessons learned from the first ACCENT training workshop in inter- and transdisciplinarity in atmospheric sciences for early career scientists. The workshop aimed at training participants in media analysis, cooperation, consensus-forming and communication skills. A real-life example, namely the burning of a waste disposal site in northern Greece, was selected as the topic of study, where unintended emissions of dioxins, PCBs, PAHs and heavy metals were encountered. The participants reported on a positive, new learning experience and expressed the wish to be offered further opportunities to learn about inter- and transdisciplinarity issues related to the atmospheric sciences.

Introduction

An academic discipline is not something that nature has given; its boundaries are historical (Mittelstrass, 1987; Wessel, 1983). Academic disciplines center around problems and areas (Stichweh, 1994), and specialize in the investigation of a specific slice of reality with a focus on particular issues. If a question or a real-world problem cannot be solved by one specific academic discipline alone, an interdisciplinary approach is sought, e.g., the coordinated and integration-oriented collaboration of researchers from different academic disciplines. The term “interdisciplinary problem-solving” originally stems from policy sciences (Pohl and Hirsch Hadorn, 2006) where problem-solving is oriented towards common interests. Today, “interdisciplinarity” is often used in a very heterogeneous way including “multi-“, “pluri-“ or “cross” disciplinarity. This order is almost always followed by transdisciplinarity, which is a specific, actor-related form of problem-oriented interdisciplinary and lets non-academic institutions and stakeholders participate in the research and production of know-how

(Pohl and Hirsch Hadorn, 2006). Although inter- and transdisciplinarity are relatively new words, the concept of taking up concrete problems of society and working out solutions in cooperation with scientists and practitioners has a long tradition. Julie Thompson Klein (1990) states that “interdisciplinarity has been variously defined in this century: as a methodology, a concept, a process, a way of thinking, a philosophy, and a reflexive ideology. It has been linked with attempts to expose the dangers of fragmentation, to re-establish old connections, to explore emerging relations, and to create new subjects adequate to handle our practical and conceptual needs. Cutting across all these theories is one recurring idea. Interdisciplinarity is a means of solving problems and answering questions that cannot be satisfactorily addressed using single methods or approaches”.

Three forces call for inter- and transdisciplinarity: (i) science that is increasingly fragmented and accountable to society, (ii) the complexity of scientific questions (e.g. air quality), and (iii) the construction of shared knowledge that is socially robust, scientifically reliable and stable (Thompson Klein et al., 2001). Inter- and transdisciplinarity can thus be viewed as a new research principle and a concept for mutual learning between experts from different academic backgrounds and with practitioners, rather than a theoretical framework. For a research network investigating atmospheric composition change such as ACCENT, it becomes crucial to move away from the classical model of knowledge production and integrate not only academic and technical, but also societal, popular and traditional forms of knowledge. Besides delivering a contribution to problem-solving and dissemination of scientific results, this new mode of research not only creates new know-how, but also “develops its own distinct theoretical structures, research methods and modes of practice, though they may not be located on the prevailing disciplinary map” (Gibbons et al., 1994). The new know-how and skills lead to communication networks in which the researchers are integrated.

The future will be even more dynamic. Scientific issues will remain difficult to understand, as the complexity of issues addressed increases (e.g., air quality-climate links). Due to the global nature of many emerging scientific issues, international collaboration that includes scientists from different cultural backgrounds will become the rule. At the same time, more diverse groups of stakeholders and lay people will emerge to whom information has to be presented in a clear and simple way so that informed judgments are possible (Brimblecombe and Schuepbach, 2006). The time when young people acquire training for a specific discipline and remain in that area to the age of retirement has passed. The economic future will belong to those who can acquire and apply the knowledge and skills demanded by the global labor market. These workers will require regular updating of their knowledge, skills and competence (lifelong learning). In a knowledge-based society, learning and teaching in higher education need to be re-fashioned towards “learning to learn”. Modular schemes and credit systems will have to be extended and deepened to provide genuine choice at various ages

and all stages of a lifetime of learning. New methods and technologies will be required to satisfy the next generation of atmospheric scientists, extending to lower graduates (e.g., Zeyer and Wyss, 2006). In conclusion, training in how to use many forms of information and knowledge, how to work in teams with experts from many different fields, and how to learn and adapt quickly and permanently to ever-changing situations are new imperatives for preparing future professionals (Thompson Klein et al., 2001).

In response to the anticipated changes in research and higher education in the 21st century, the ACCENT Task “Training and Education” has outlined its objectives, strategies and plans for 2004-09 in a Position Paper (Schuepbach, 2005). Among the defined objectives are the development of integrated learning environments and implementation of “best practice” in using a Network of Excellence for the training of the next generation of atmospheric scientists. Besides scientific training, early career scientists are offered training in soft skills, outreach to the general public, inter- and transdisciplinary collaboration, and the use of the New Information and Communication Technologies (NICT) for learning, teaching, and collaborating. The ACCENT Network can draw on a pool of competences in all areas. Partners in the Network contribute to the training workshops with the know-how and tools from their academic discipline and scientific fields (scientific training). This allows the early career scientists to learn, under the guidance of ACCENT scientists, how a problem is perceived, investigated and managed in different ways (see also Defila et al., 2006). Here, we report on a training workshop on inter- and transdisciplinarity organised by the T&E Task group in the frame of a workshop on “Air Quality in the Mediterranean for the Next Generation”, held in Greece from 9-12 October 2006.

Objectives of the Training Workshop

The world faces huge challenges in improving the quality of life by reducing air pollution. Emissions involved stem from individual traffic, the quality of cars and motorcycles, public transport, and the behavior of citizens (e.g., waste disposal). A sustainable improvement in air quality and thus in the quality of life can only be achieved if representatives of all interest groups work together: politicians have to develop strategies, health professionals need to assess impacts, communication channels (e.g., media) need to be utilised to inform the general public, etc. Leaders of an air pollution management project must have the necessary know-how, instruments, intervention techniques and skills, jointly with the scientific experts, to bring the program to a success. Among these skills are techniques to promote self-organizing processes, application of effective means of communication, and forms of conflict resolution to address issues of diverging interests.

In view of these challenges, a training workshop on inter- and transdisciplinarity was designed as part of a suite of training workshops on “Soft and Essential Skills” in T&E of ACCENT. A real-life example was selected, that being the burning of a waste disposal site at Tagarades

near Thessaloniki in northern Greece. The fire broke out on 14 July 2006 and lasted for two weeks; approximately 80,000 tonnes of waste was burnt. Three research groups from the Aristotle University of Thessaloniki were asked to investigate the extent and severity of the air pollution emitted by the fire. Approximately 50 g (I-TEq) of dioxin were reported to have been emitted into the atmosphere of the surrounding agriculture land. Samples (soil, grass, ash, milk, eggs and other agricultural products) were taken and analyzed (PCB, PAH and heavy metals) by different institutions in Greece. The burning event and subsequent scientific study was a focus of public interest, even more so as elections were held in Greece in mid-October 2006.

The major aspects to be studied in the training workshop were (i) the nature of interdisciplinary collaboration among the scientists involved in the sampling and analysis, (ii) the degree of transdisciplinary management of the accident, (iii) clarity and timing of information to the public by responsible bodies, and (iv) the role of the media and interaction with the policy makers.

Procedure, Methods and Lesson Plan

Presentations by external experts

Five scientists and stakeholders were invited to present their views and findings on the fire at the waste disposal site from the perspective of an engineer (Prof. N. Moussiopoulos), a chemist (Prof. K. Samara), an epidemiologist (Prof. H. Tsoukali), an air quality manager (Dr. K. Nikolaou), and a conscious citizen (Mr. S. Famellos). Their short presentations in the beginning of the training workshop were given in Greek with simultaneous translation into English. After the presentations, the participating early career scientists asked questions and then withdrew for group work.

Workshop participants and working groups

The twenty-five early career scientists participating in the 1-day training workshop originated from thirteen countries and had different scientific backgrounds (geology, oceanography, environmental engineering, atmospheric sciences, remote sensing). In order to simulate real-life experience, they were split into four groups and asked to work on specific topics as outlined below. The expected outcomes of the group work were a brief oral presentation for the plenary session held in the late afternoon and a one-page handout for feedback by peers the following morning. Each group had about six members and was chaired by a senior or experienced scientist; no instructions were given to the chairs as to how to lead their groups.

Only Greek participants were accommodated in Group 1 (Media Analysis), chaired by Prof. Peter Brimblecombe, University of East Anglia, Norwich, U.K. Their task was to analyse content, icons, and pictures of about 200 compiled newspaper articles on the burning event with a view on health and safety versus air pollution regulations. Group 2 (Policy Briefing) was chaired by Dr. Arnolds Übelis, University of Latvia in Riga, and aimed at producing a briefing paper on the burning event for politicians,

similar to the UK Parliament Post Briefing. Group 3 (New Direction Article) had to write a scientific summary paper on the burning event for other scientists, based on the New Direction Articles in Atmospheric Environment. The group was chaired by Mark Jacob, University of Freiberg, Germany. Group 4 (Interdisciplinary Disagreement) was chaired by Dr. Evi Schuepbach, University of Berne, Switzerland. It explored the underlying scientific models and potential disagreements in the presentations given by the experts and stakeholder in the morning of the training workshop.

The plenary session in the afternoon assembled all participants and work group chairs. The feedback the following morning merged Group 1 with Group 3, and Group 2 with Group 4. A technique called “fishbone” was employed to offer feedback to the authors of the handouts. The feedback round was preceded by an analysis of the group work (i.e., reflecting on roles in the team, leadership, how difficulties were handled, etc.), an evaluation of the entire workshop, and the introduction of the fishbone technique.

Outcome of the Working Groups

Group 1: Media Analysis

The media interest in the burning event lasted about two weeks, standing in contrast to the time-scale of political action of years. Most popular newspapers published frightening pictures of the event and used strong words (e.g., toxic bomb, dead zone, nightmare, etc.). The information disseminated to the general public was mostly qualitative in nature; quantitative information was sparse and not precise. According to the media, the local authorities, the government and policy makers were responsible for the burning event. No personal consequences were requested. In summary, the media frightened people more than promoting social change and awareness (e.g., recycling), and focused more on the (political) problem than on (social) solutions.

Group 2: Policy Briefing

The collective knowledge of dioxin goes back to the mid 1980s, with the most recent international scientific conference on dioxin organised in Oslo, Norway, from 21 - 25 August 2006. The published material in the open literature on dioxin provides evidence that knowledge about the danger of dioxins in waste management exists. A campaign to engage the public is viewed as an important part of a strategy to reduce emissions of dioxins and dioxin-like PCBs from fires. Such engagement would need to be combined with other national and local measures to encourage and enable members of the public to play their part in reducing emissions. Three recommendations as isolated from the published literature can be formulated: 1. Don't burn household waste indoors or outdoors, and especially not waste containing plastic; 2. For people who have coal fires, choose coal with a low chlorine content, and make sure the fire burns efficiently, and 3. Don't burn waste on bonfires – in particular, avoid burning plastics.

Group 3: New Directions

Because of the time it took the responsible authorities to respond to the fire (~5 days), measurements of PM10, dioxins, furans and heavy metals released by the fire were only measured a week after the fire had started, when the estimated daily amount of waste burnt was an order of magnitude lower than at the start of the fire. The concentrations of lead and cadmium measured in edible substances such as milk, vegetables and fruit close to the burning site thus were, with the exception of one sample site very close to the fire, all under the recommended health safety limit. Although such data are useful in terms of assessing health impacts on the local population, crucial data from the first week of the fire is not available. A different approach to problem-solving is recommended in the future, e.g., closer cooperation between policy makers, scientists and landfill operators (and stakeholders in general) for quick and adequate measurement and model reactions to such incidents. This cooperation could be at three different levels: planning, monitoring and emergency response.

Group 4: Interdisciplinary Disagreement

The scientific tools and methodologies used to investigate the burning event were (i) dispersion modelling, (ii) waste and risk management, (iii) toxicology (pathway ingestion), and (iv) analytic sampling (pathway air). The conceptual model for waste and risk management is a chain of research from dispersion modelling to sampling (based on the model results); the communication of the information to the general public; prevention – i.e. site assessment (chemical and toxicological analysis); tuned models for specific areas; and the definition of specific scenarios for potential accidents. The same conceptual framework was found to underlie the field of analytical chemistry and toxicology. The analysis of the expert presentations indicated that the research had been carried out separately by the individual scientists involved in the investigation of the waste burnt; there was no interdisciplinary collaboration. Yet, there was also no disagreement between the disciplines for the final recommendations, and the scientists reached similar conclusion, i.e.: the health impact was within the limit of what is accepted by the World Health Organisation (WHO).

Workshop Evaluation

Were the learning goals achieved? For the evaluation of the workshop, the participants were introduced to a method called “fishbone”. This method let one group sit in an inner circle and evaluate the workshop, while the other group assembled around the inner group and listened. The groups then swap. This process resulted in an analysis of the dynamics during the group work, including reflections on team work and leadership, and of the workshop as a whole. Questionnaires were also used to assess the workshop. The summary of feedbacks shows that the experiences of the early career scientists were different in each working group. Most participants indicated that the training brought new ideas and was a good practical exercise. Understanding of the topic

was felt to be advanced by the presentation of different perspectives on the subject given by the experts and stakeholders. However, a more detailed perception from a politician would have been appreciated. For some, the waste disposal case study was too specific as a topic, albeit seen as interesting from the perspective of risk management, controlled waste and environmental accidents. It was proposed to have a more general topic on atmospheric sciences included in a next training on inter- and transdisciplinarity. It was also noted that sustainable development should be integrated in the discussion, particularly with regard to assessing new strategies.

Many participants reported have a positive, new learning experience in structured working and how a group can quickly come to a result, given a very strict schedule. The fact that a group with different scientific backgrounds and from different cultures was able to interact and exchange on a new topic (waste management) was considered rewarding. The training workshop also made some participants appreciate the value of communicating and maintaining a dialogue, and it raised awareness of the difficulties in communicating and collaborating in the context of tight time constraints. It was suggested that (intercultural) dialogue, team work, time management, handling difficult situations, and gender issues should be more emphasised in future training workshops. Also, in-depth theoretical frameworks on team building, leadership and management would be welcomed in order to become familiar with the respective vocabulary. For many, the changing role they took in the group as work progressed was a new and satisfying experience. Being asked what the major challenges are in acting as a leader, some participants said it is the coordination of different tasks and strong-minded scientists. Others expressed anxieties about assigning responsibilities at all, or having to assign them relative to the skills of each individual so as not to offend anyone. Establishing a common, acceptable time schedule, the synchronization of work and a clear definition of the expected outcome of a task was also seen as difficult. As a conclusion, more training in leadership, conflict resolution (mediation) and time management would be appreciated, in view of the cultural diversity inherent in global collaboration.

The role of the work group chairs was very variable, ranging from carefully giving directions where the group needed it or asked for it, to interactive participation in the group work or even leading the group. Some participants consequently enjoyed peer-learning, while others felt rather frustrated in the beginning of the process because interaction in the group was not encouraged. There were some negative feedbacks on the infrastructure: the rooms were mostly not perceived as suitable for group work and a lack of computers and access to the Internet was reported. Interestingly, none of the participants would have liked to receive the training material prior to the training workshop in order to get prepared.

Conclusions and Further Avenues

The ACCENT training workshop on inter- and transdisciplinarity offered an opportunity for a group of early career scientists to analyse and experience how an

environmental issue was handled in real life. During this learning process, they became familiar with discipline-specific approaches to problem-solving that may be different from their own disciplinary background. They had to integrate knowledge and the vocabulary of other disciplines while at the same time contributing as a specialist and reflecting on their own approach. By trying to find a common language and view of the problem, the early career scientists learned to set their discipline in relation to other disciplines. This process created new skills and know-how, and led to a new network. The wish was expressed that more opportunities to learn how to work in an interdisciplinary, multicultural team should be offered, alongside training in cross-cultural dialogue, conflict resolution and gender issues. The Task T&E in ACCENT will attempt to comply with these needs, and it also will aim at involving more social scientists in future training workshops to accompany the groups such as in an experiment in the USA (Haag, 2006). Analysis of all available multimedia documentation (digital video, photographs) produced during the training workshop will offer additional insights.

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Modeling of Deep Convection and of Chemistry and their Roles in the Tropical Tropopause Layer: SPARC-GEWEX/GCSS-IGAC Workshop: Victoria, BC, Canada, June 12-15, 2006

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Introduction

The Tropical Tropopause Layer (TTL) is an excellent example of coupling between dynamics, radiation (including cloud feedbacks) and microphysics. Convection in the tropics plays a key role in redistributing trace gases and aerosols in the upper troposphere-lower stratosphere (UTLS) region, and the interaction between cloud dynamics, radiation and microphysics is of fundamental importance in these processes. The global scale models used to study climate typically have spatial and temporal resolutions that limit their ability to properly represent many of these processes, yet a solid understanding of their effects is necessary. Cloud Resolving Models (CRMs), particularly when supported by appropriate observations, can be used to better understand the different processes and their interactions.

The TTL has received much attention within the WCRP-SPARC¹ community because of its strong link with stratospheric climate. Among the many processes operating within the TTL are those that control the amount of water vapor and trace gases entering the UTLS, important in determining greenhouse gas forcing in the tropics, and determining stratospheric chemistry

¹ World Climate Research Program's Stratospheric Processes and their Role in Climate project (<http://www.atmosph.physics.utoronto.ca/SPARC/index.html>)

and temperatures through the formation of the hydroxyl radical (OH), polar stratospheric clouds (PSCs) and enhanced radiative cooling.

Within the IGAC community there is considerable interest and active research in regard to the role of deep convection in transporting and processing chemical constituents and aerosols. The WCRP/IGBP Atmospheric Chemistry & Climate Initiative (AC&C), currently being developed under the joint leadership of SPARC and IGAC has the objectives of understanding and quantifying the processes that link surface emissions of constituents and precursors to atmospheric composition, and how this composition and its changes are in turn related to climate forcing and change. Modeling is a key initial component of this initiative and can address a number of issues including the identification of processes that are associated with the most uncertainty in radiative forcing and climate change, the development of metrics that provide insight into modeling of chemistry-climate interactions on a range of spatial and temporal scales, the identification of more (or less) promising approaches to representing key processes in climate models, and can contribute to the identification opportunities for programs and projects whose goal is to improve climate models and our understanding. In this context, the TTL is of particular interest.

Research on modeling and understanding of deep convection in the tropics has also received considerable attention within the WCRP-GEWEX² community. The goal of the GEWEX Cloud Systems Study (GCSS) is to support the development of cloud processes in climate and forecast models. This is accomplished by: developing the scientific basis for the parameterization of cloud processes; coordinating the acquisition and assimilation of observations; coordinating the use of CRMs to support the development of parameterization schemes in large-scale models; and promoting the evaluation and intercomparison of parameterization schemes for cloud processes.

This workshop brought together expertise from these communities, and related research activities on stratospheric processes and modeling of deep convection and chemistry in the tropics to set the stage for a better understanding of the role of convection in determining the thermal structure and composition of the TTL. The workshop involved plenary sessions, poster presentations, two sets of breakout periods in the afternoon of the third day, and final summary and discussion periods on the last half-day where future directions and actions were proposed and discussed. The plenary sessions were organized along three themes: Overviews of historical development and current TTL research (Day 1); Modeling deep convection in the tropics (Day 2); and Coupling of deep convection and chemistry (Day 3). The two very well-attended poster sessions were highly complementary to the oral sessions and served to highlight new research efforts and directions. This brief report summarizes key aspects of the scientific presentations, the outstanding questions and issues brought forward in breakout

² World Climate Research Program's Global Energy and Water Cycle Experiment (<http://www.gewex.org/>)

Tropical Tropopause Layer and Deep Convection

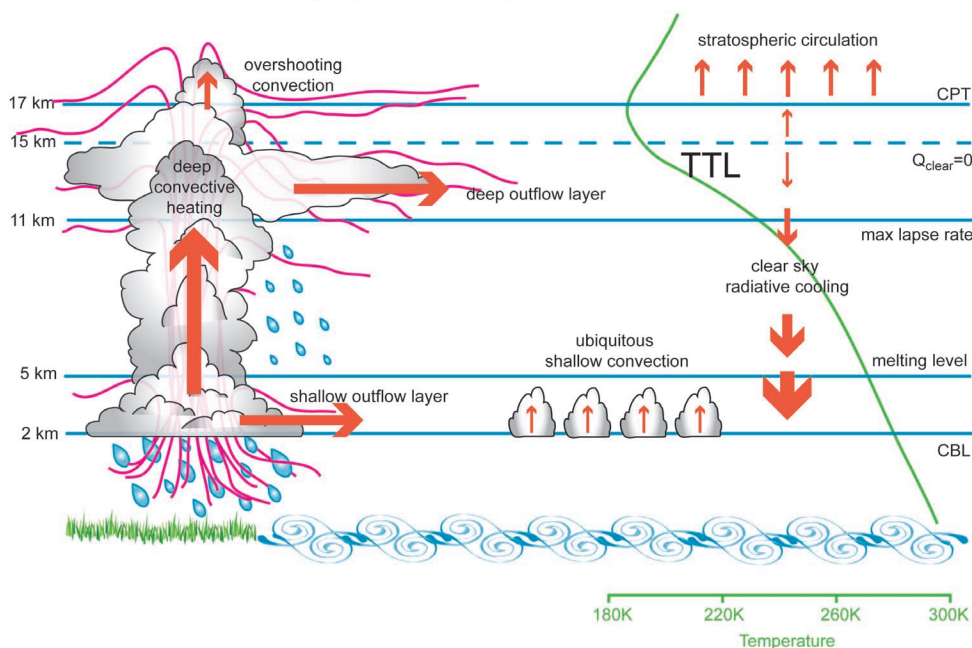


Figure 1. A pictorial view of the Tropical Tropopause Layer (TTL) and deep convective transport.

discussions, and the actions proposed in the subsequent discussion periods.

Overview of current directions in TTL research

Research in the last decade has served to establish the nature of the TTL as a transition region in which the air has mixed stratospheric and tropospheric properties, which in turn are determined by a combination of both large-scale and convective processes. Elucidating these processes has motivated a substantial amount of research in recent years. The overall aim of the invited oral presentations on the first day was to review this research, summarize the knowledge that has been gained and provide insight into outstanding questions. These presentations, in combination with the first poster session, effectively set the stage for the more focused attention on modeling convection and chemistry in the following two days.

The vertical structure and some salient properties of the TTL were discussed in the overview presentations by I. Folkins and A. Gettelman. Figure 1 illustrates some of these properties.

Observations in the tropics indicate a tri-modal structure of cloud tops associated with convection (I. Folkins). In addition to deep convection with cloud tops in the upper troposphere and shallow trade-wind boundary layer cumulus, cumulus congestus clouds with tops typically in the range of 5 km above sea level (ASL) are ubiquitous in the tropics. In regions where convection plays an important role in the time-averaged large-scale energy and moisture budgets, there is typically a close balance between the net downward mass flux associated with the

large-scale vertical motion in the clear areas surrounding convective regions and the net upward flux associated with convective scale motions in the enclosed cloudy areas. The radiative cooling in these clear areas in turn nearly balances the heating associated with the large-scale mean vertical motion. Thus important features of the mean convective scale mass flux profile can be estimated using sounding data in clear areas. Such profiles reveal regions of deep convective outflow in the upper troposphere, typically between 10 km and 18 km with a local maximum typically near 12-14 km ASL. The level at which clear-sky net radiative heating vanishes ($Q_{clr} = 0$) is located near the top of the convective outflow layer but a significant amount of convection penetrates above this level.

Other defining features of the TTL region include a maximum in the temperature lapse rate in the upper troposphere, often found near the level of maximum convective outflow, and a less pronounced minimum in ozone near this level with a rapid increase above, and a more uniform vertical structure below (A. Gettelman). To first order the transition from a more uniform distribution of ozone in the troposphere to the rapidly increasing profile in the TTL is evidence for the reduced vertical mixing efficiency in that region relative to that which occurs within the tropical troposphere. However the presence of an ozone minimum may be the result of the outflow of ozone-depleted air which is transported upward from the boundary layer in deep convection.

The cold point tropopause marks a sharp transition in the vertical temperature gradient (i.e. in static stability, poster by T. Birner). Although several definitions of the TTL have been proposed, a useful thermodynamic one includes the region between the maximum in the temperature lapse rate and the cold point (A. Gettelman). In view of the small vertical scales associated with the structure of salient features of the TTL, the question of whether this region can be adequately represented in Atmospheric GCMs (AGCMs) is of practical importance. However, notwithstanding the limitations imposed by the vertical resolutions of most current AGCMs there is some evidence that these models are able to simulate some of the main features of the TTL in a realistic manner (talk and poster by A. Gettelman and poster by T. Birner).

The clear-sky component of the radiative heating in the TTL region above the level of $Q_{clr}=0$ has both longwave and shortwave components, but it is mainly associated with

shortwave absorption by CO₂ (predominantly), O₃, and H₂O. Contributions to net radiative heating in this region may also come from elevated cloud layers. In the zonal mean the radiative heating in the upper part of the TTL is partially offset by cooling associated with ascent in the Brewer-Dobson circulation which, though predominantly located in the stratosphere, extends down into the TTL. However the TTL has a 3-dimensional structure and the ascent rates required to offset the radiative heating are in general larger than those associated with the Brewer-Dobson circulation (noted in the presentation by S. Sherwood). In the TTL other dynamical processes that play important roles include large-scale horizontal transport and the influence of tropical waves on various scales which may propagate into the region and, among other things, produce localized temperature and constituent fluctuations (discussed in the presentation by W. Randel). The temperature variance associated with these waves shows a maximum close the cold point (also W. Randel).

The magnitude and variation of water vapor in the tropical lower stratosphere is regulated within the TTL. The minimum in water vapor that is typically found in the stratosphere near the tropopause is indicative of dehydration of air ascending into the lower stratosphere. Elucidation of the processes that are responsible for this dehydration is currently the subject of active research and was discussed in several of the presentations given at the workshop. Dehydration associated with large-scale transport is a mechanism that has been explored extensively in the context of Lagrangian trajectory studies (illustrated in presentations by P. Haynes and L. Pfister), wherein possible trajectories of air parcels passing from the troposphere to the stratosphere through the TTL are determined using wind fields derived from large-scale reanalyses. In this mechanism, the net dehydration of a parcel that enters the stratosphere is determined by the coldest temperatures it encounters during the lifetime of its trajectory (the Lagrangian cold point). Thus although dehydration is associated with large-scale transport and uplifting, the three-dimensional structure of the TTL is important in this mechanism. This in turn may be affected by overshooting convection in the TTL. The results obtained in recent trajectory studies support the conclusion that stratospheric water vapor is largely controlled by large-scale transport (P. Haynes). The introduction of convective overshoots into the trajectories had an overall hydrating effect (L. Pfister). One remaining problem in these trajectory calculations is the lack of high supersaturations in the TTL, as found by recent aircraft measurements. Explaining the physical processes that lead to high levels of supersaturation in upper tropospheric ice clouds is a current research challenge. Nevertheless, even rare convective overshoots may have a significant effect on constituent transport into the TTL and lower stratosphere. S. Sherwood estimated that a convective cloud fraction of as little as 0.02% is sufficient to matter. Furthermore, he presented evidence that convective overshoots lower and cool the cold point, thus affecting temperatures in the TTL.

The conclusion that to first order stratospheric water

vapor can be explained by large-scale processes does not preclude the possibility that it may be directly affected by convective processes. A plausible mechanism of dehydration could be associated with overshooting convection. Such overshooting is inevitably accompanied by entrainment of environmental air into turrets, followed by gravitational settling and mixing with the environment. If a substantial amount of the ice produced by the cooling and condensation associated with the overshooting falls out of the TTL before being detrained and mixed into the environment this process will give rise to dehydration. The opposite effect (hydration) may occur if the ice is detrained and evaporates in the process of mixing. Strong evidence for moistening via this process has been found recently from aircraft measurements indicating the presence of ice particles above the tropopause close to deep convection in the TROCCINOX and SCOUT-O3 campaigns (presentation by T. Corti). The roles of microphysical processes and other constituents in the TTL and hydration/dehydration are currently the subjects of active research, aspects of which were presented in both oral presentations and a number of the posters.

There is currently some debate concerning the possible role of convection in the thermal structure and heat balance of the TTL. This is illustrated by the recent studies of Kuang and Bretherton (2004) and Küpper et al. (2004) who used CRMs in similarly designed idealized experiments exploring a quasi-equilibrium model involving a balance between radiative heating and cooling by convective overshooting and (imposed) large-scale ascent. However they have reached different conclusions (oral presentation by C. Bretherton). Kuang and Bretherton concluded that radiative heating is predominantly balanced by convective cooling, while the conclusion of Küpper is that the convective cooling contribution is relatively small. The reasons for these differing conclusions are currently not fully understood.

Putting this in the context of observations, W. Rossow gave a survey of available satellite measurements of tropical convection. The top of convection from these measurements can be deduced from brightness temperatures in relation to the cold point temperature, and therefore systems penetrating the tropical stratosphere can be inferred. He showed that larger systems are more likely to contain penetrators and that these preferentially occur during the early stage of the lifetime of the convective system.

Modeling of deep convection and its role in the TTL

The session on modeling of deep convection and its role on the TTL aimed to provide an overview of the current research being done by those involved in the GCSS “Deep” working group (formally, the Precipitating Convective Cloud Systems WG). Besides a range of posters presented over the week there were several invited talks in which each speaker was asked to discuss a specific topic and describe how this work related to the issue of the TTL. The goal of these talks was to provide the workshop with details of the current state-of-the-art research into

convection and to enable attendees to consider how this can be linked to problems with modeling the TTL.

The session began with J. Petch, the chair of the GCSS “Deep” WG, who discussed the recent work carried out by this working group. This presentation focused on a multi-model intercomparison of simulations of the Tropical West Pacific during TOGA-COARE. Various model types were involved in this study including Cloud Resolving Models (CRMs), single column models (SCMs), numerical weather prediction (NWP) models, and climate models run in NWP mode. An analysis of the behavior of the models in the upper troposphere highlighted significant differences between the different model types. Deficiencies in the current experimental designs for the CRMs and SCMs for studying the TTL were discussed with suggestions for improvements.

Two talks covered aspects of observations and model evaluation. E. Zipser discussed microphysics observations in the tropics and how these can be used to evaluate CRM simulations. Comparing a CRM simulation of a mesoscale convective system with observations made during the KWAJEX field program, he showed that the CRM tended to overestimate convective intensity somewhat, and graupel mixing ratios considerably. If an error in CRMs of the size shown is typical, it suggests that this would have significant implications for the use of CRMs to study transport in the TTL. He was keen to stress the difficulty and importance of assessing CRMs with observations and suggested that from the perspective of an observer of “the real world”, attempting to evaluate imperfect models with imperfect data, and draw conclusions from this exercise will probably be “imperfect, squared”. A further talk about available observations for the evaluation of models was presented by the chair of GCSS, C. Jacob. He described the Tropical Warm Pool International Cloud Experiment (TWP-ICE) which took place in Darwin in tropical North Australia during January/February 2006. The aim of the experiment was to study tropical cloud systems and their environment during monsoonal conditions in a holistic way. Implications for modeling, including the derivation of suitable forcing and validation data sets for Cloud-System-Resolving Models and Single Column Models, was also discussed.

The current state-of-the-art development in convective parameterizations was presented by L. Donner. It was stressed that with the emergence of Earth system models there are substantial new requirements for parameterizing convection. In addition to their traditional roles in providing heat and moisture sources and sinks for large-scale atmospheric flows, convective parameterizations must now treat tracer transport, in-cloud chemistry, and scavenging processes. Also, because of indirect effects on clouds by aerosols, both warm and cold microphysics must be considered within a deep convection scheme. In the latest developments of his scheme, he showed the importance of more realistic treatment of cumulus-scale vertical velocities, explicit incorporation of mesoscale circulations associated with deep convection and new insights on closures for cumulus parameterizations. The importance of the generation of upper-tropospheric ice,

injection by deep convection of tracers into the stratosphere, and impact of convection on tropical transients was also discussed. In a related talk, D. Williamson went on to describe how parameterizations within a climate model can be tested using NWP type simulations. Short forecast errors and the balance of terms in the moisture and temperature prediction equations, which lead to errors, were compared at the ARM (Atmospheric Radiation Measurement) Southern Great Plains site for the June/July 1997 and April 1997 Intensive Observing Periods. Errors seen in the NWP forecast runs can often be linked to those seen in climate runs.

Two presentations discussed the concept of the “super-parameterization” – the use of embedded CRMs into each grid box of a larger scale model to represent a number of the sub-grid processes. D. Randall presented an analysis of an AMIP³ run with the Super-CAM⁴ showing a range of benefits to this technique, particularly in terms of capturing intra-seasonal, seasonal, and interannual variability. W. Grabowski, the pioneer of this technique, then went on to discuss how a similar method could be applied to a mesoscale model. He discussed which processes would be resolved by which model and suggested the relevance of this for modeling tropospheric-stratospheric coupling. In a related talk, G. Shutts showed how a very large domain CRM had been used to study the interaction between deep convection and large-scale tropical flow. The model was configured with equatorial beta-plane geometry and used a highly anisotropic grid in the horizontal to reduce the computational burden. Characteristics of the tropical tropopause layer in the model were discussed as well as the different tropical waves produced in the model. As with the GCSS case, there were some problems in the experimental design for study of the TTL but suggestions were made on how these could be addressed.

Three talks focused on different aspects of cloud resolving modeling and how it related to the TTL. P. Yau provided a general discussion about simulations with a high resolution model during TOGA-COARE which suggested there is notable sensitivity to resolution, with many different scales important in the convection. E. Jensen described the use of bin-microphysics in modeling deep convection and of its importance when considering the role of microphysics in the TTL. He showed that the suggested mechanism, where extremely cold air generated in overshooting deep convection can provide a significant source of dry air to the TTL, did not occur in their simulations of updrafts penetrating the tropical tropopause. This was because the mass of ice was in relatively small crystals which did not precipitate out sufficiently. However, for a different simulation using a CRM with bulk microphysics, D. Grosvenor did show that overshooting convection during TOGA-COARE could lead to a significant reduction in the total water content in the TTL.

³ AMIP = Atmospheric Model Intercomparison Project, a standard experimental protocol for global atmospheric general circulation models (AGCMs). (<http://www-pcmdi.llnl.gov/projects/amip/index.php>)

⁴ CAM is NCAR’s Community Atmosphere Model. In the Super-CAM, a Cloud System Resolving Model (CSR) has been installed as a “super-parameterization” inside the CAM.

Chemistry and deep convection

The aim of the session on modeling chemistry and deep convection in the TTL was to provide an overview of current research on the topic, to describe a collaborative study that addressed mid-latitude convection, and to discuss recent and upcoming observation programs that could be used for future collaborative studies.

The overview of chemistry in the TTL, given by M. Lawrence, covered four topics: 1) low-ozone air masses in the TTL; 2) deep convective transport of tracers; 3) scavenging of tracers especially by ice and its role for HNO_3 ; and 4) extended horizontal observations of a suite of gases (ACCENT results). The challenges of observing and modeling chemical species in the TTL were introduced. One key observed phenomenon of the TTL is extremely low-ozone air masses which seems to be influenced by deep convective transport, but other factors, such as reactions on ice, marine boundary layer halogen chemistry, and NO_x production from lightning, may also play an important role in suppressing ozone concentrations. The role of transport processes (e.g. deep convective transport), scavenging of gases by cloud particles (especially ice), slow upwelling transport in the TTL, and exchange with the stratosphere, have all been found to be important factors in determining the concentrations of chemical species in the TTL.

Simulations of tropical convection and chemistry show distinct differences over the oceans versus over continents. The organization and intensity of the convection over land allows for more convection to reach higher altitudes with overshooting turrets that reach into the lower stratosphere. The higher lightning frequency in these continental storms also results in more NO_x in the TTL. These NO_x -rich air masses chemically age while being transported to oceanic regions, resulting in air masses with increased ozone in the TTL. In addition, the surface emissions over land and ocean are vastly different. Continental emissions include a wide variety of non-methane hydrocarbons and volatile organic compounds, in addition to surface NO_x and carbon monoxide. Oceanic emissions include dimethyl sulfide, methyl iodide, and other very short-lived halocarbons, as well as sea-salt aerosols. C. Mari pointed out the importance and interaction of the diurnal cycles of oceanic convection and photochemical activity. Convection tends to peak during the nocturnal, early morning while photochemical activity is highest during the middle of the day. In severe, continental convection, E. Riviere pointed out the positive role of wave breaking on the ozone budget in the TTL. Both Mari and Riviere suggested the need for high resolution simulations of convection and chemistry, as well as for more measurements of chemical constituents near convection.

The effect of aerosols on deep convective clouds and the liquid and ice microphysical processes therein have recently become a topic of prevalent research. C. Wang presented results from his 3-dimensional convective-scale simulations of tropical oceanic convection with varying initial cloud condensation nuclei (CCN) concentrations. His four hour simulations showed the non-monotonic

response of various cloud properties to increasing CCN and that dynamics and ice microphysics both play important roles in altering these properties.

In looking toward possible future activities that could be done, M. Barth showed results from an intercomparison of models simulating mid-latitude deep convection and chemistry. The models participating in the exercise agreed quite well with each other, and with measurements of the passive tracers CO and O_3 . There was fairly good agreement between measurements of NO_x , which was produced primarily from lightning, but there was less agreement between measurements of soluble species, such as hydrogen peroxide, formaldehyde, and nitric acid.

Evaluating these model simulations and understanding the role of deep convection and chemistry on the TTL would require a suite of chemical, physical, and dynamical measurements in and near deep convective storms. K. Pickering outlined the recent and upcoming field experiments of CRYSTAL-FACE, TROCCINOX, SCOUT-O3, ACTIVE, CR-AVE, AMMA, and TC4. Of the field experiments occurring before June 2006, the measurements of cloud physics, passive tracers (CO and O_3), NO_x , and lightning were quite good. It was suggested that CRYSTAL-FACE and TROCCINOX sampled primarily subtropical convection although one excellent tropical event was sampled during TROCCINOX. There were several days of sampling the HECTOR storm during SCOUT-O3 and ACTIVE and several events in contrasting regimes sampled during ACTIVE and TWP-ICE. Thus, it is promising that a tropical deep convection case can be used for future modeling studies of chemistry and thunderstorms.

Key questions and issues

Two sets of breakout sessions were held on the afternoon of the third day of the workshop with a short plenary between them to air questions and discussion points from the first set of breakouts and which would be considered in the second set. The first set of three breakout sessions were organized along thematic lines, with one devoted to a discussion of GCSS Deep Convection Working Group case studies, a second to chemistry issues in the TTL and a third to large-scale processes in the TTL. The overarching aims of the breakout sessions were to (a) assess current knowledge and understanding, (b) identify questions and uncertainties that could be addressed through future cross-cutting and cross-programmatic collaborations, and (c) propose ways of addressing these questions and issues.

The chemistry breakout session discussed the importance of convection on chemistry with three major points. First, convection can affect the ozone production efficiency ($k[\text{NO}][\text{HO}_2]$) by supplying HO_x precursors to the TTL and by supplying NO_x (via lightning production) and removing NO_x (via HNO_3 uptake). Second, convection can transport Very Short Lived (VSL) halogens to the TTL, potentially altering global tropospheric photochemistry and the stratospheric halogen boundary condition. Third, while convection detraining above $Q_{\text{clr}}=0$ affects the

composition of the TTL the most, convection detraining at lower altitudes can affect the global troposphere.

The large-scale processes breakout session discussed a number of issues. One of the outstanding problems concerning trajectory calculations is that the transport and life times appear to be wrong. These errors may be related to the absence of mesoscale dynamics (e.g. gravity waves, mesoscale convective systems) in the meteorological analysis fields. In addition, the absence of turbulent mixing in trajectory calculations may add some uncertainty to these Lagrangian analyses. A second issue discussed was the ability of CRMs to capture the interaction between the cloud scale and large scale given the small domain sizes used in CRMs. A third point brought forward was that it may be difficult to use tracers to distinguish between two dynamical processes, i.e. convective detraining and slow uplifting, which could produce very similar tracer distributions in the TTL.

Several key questions emerged from the first set of breakout sessions:

1. How accurate are CRMs in representing key cloud-scale processes in the TTL?
2. What is the role of overshooting deep convection in setting the cold point (CP) temperature?
3. What is the role of convection in determining ozone and ozone depleting substances (ODS) in the TTL, and therefore in determining boundary conditions for ODS in the stratosphere?
4. How well do CRMs model convective outflow and the distributions (or PDFs) of detraining?

The second set of breakout sessions discussed possible means of addressing these questions and identified some modeling and observational approaches that would be necessary and/or desirable to succeed in this objective.

The accuracy of CRMs can be addressed via intercomparisons between models and validation via observations. In this regard it will be useful to utilize the framework of GCSS which involves comparisons of several models, all of which are run for a particular case where good forcing data and observations are available; the benefits of this methodology have been well documented within GCSS. In order to evaluate both the role of convection in the TTL and the ability of models to account for it in a physically realistic way it will be necessary to include additional processes and additional observations. This is where active collaboration of experts from IGAC (chemistry) and (trop-strat exchange) SPARC communities will be critical for success. For example the use of chemical tracers such as CO and O₃ must be part of these studies.

Appropriate forcing data sets for CRMs, satellite data, and large-scale models that represent the coupling between convective and larger-scale circulations are all needed to address questions regarding the role of convection in determining ozone and ozone depleting substances (ODS) in the TTL. Mass flux analyses should be part of future intercomparison studies. A field study (similar to the

previously proposed TTL-Chem), conducted in concert with cloud-scale and large-scale model simulations would be valuable to quantify the mass flux of key compounds from convection into the TTL.

Future directions and activities

A number of suggestions for future activities and collaborations resulted from the summary of break-out sessions and the ensuing discussion on the last day of the workshop. Several of the immediate and very near term actions suggested have been carried out, are under way, or planned:

- Posting of workshop presentations on the SPARC web site (See http://www.atmosp.physics.utoronto.ca/SPARC/TTL/Participants_Presentations.html)
- Posting of a bibliography of TTL and related literature on the SPARC web site (currently being assembled)
- Posting a list a field experiments (from K. Pickering and C. Jakob's talks) and identify contact people from these experiments for model analysis.
- Solicitation and publication of summary articles in the SPARC, IGAC and GEWEX newsletters on scientific topics and results discussed in the workshop and other related work.

Suggestions for the intermediate term (1-2 years) included organizing special sessions on the role of convection in the TTL in upcoming scientific meetings and holding follow-on workshops (in 18 months to 2 years) that may deal more exclusively with some key issues that are of importance in understanding the role of convection in the TTL. These could possibly focus on results of recent field campaigns and efforts to use the results to constrain CRMs. For example observation and modeling of short-lived chemical constituents may provide insight and constraints on convective mass flux distributions in the TTL.

In regard to coordinated modeling activities, it was suggested that it may be possible to revisit some of the case studies that have been used by the GCSS Deep Convective Working group (e.g. TOGA COARE) in previous CRM intercomparisons with the objective of better understanding of issues in modeling of convective scale processes of importance in the TTL. For example, an initial step could be to repeat the 2006 GCSS Deep Convection Intercomparison case of TOGA-COARE with the inclusion of 1-2 tracers. Activities are already under way (talk by P. Yau) using a multi-grid modeling approach in the context of TOGA-COARE to study the role of convection in the TTL. Future modeling intercomparison case studies to facilitate examination of TTL processes could include observations from more recent field campaigns such as TWP-ICE (talk by C. Jakob), TROCCINOX, SCOUT-O3, ACTIVE, (talk by K. Pickering) and AMMA, which took place August 2006.

It was widely agreed that an important first step toward establishing a framework for future collaborative activities is to compose a working group to build on the groundwork laid down by the workshop. This group

should be comprised of researchers in the three broad communities that participated in the workshop. The working group will develop a framework for further collaborative research and plan future activities. A number of possible members for the working group were suggested. The workshop organizing committee has taken on the task of bringing about this next step. It is anticipated that a small working group will correspond by email and/or meet on an ad-hoc basis over the coming year to develop plans for a collaborative program that can be proposed to the IGAC, GEWEX, and SPARC projects for the endorsement that will facilitate support for future activities. At the meeting it was agreed that initially this

group would include the workshop organizers (T. Birner, J. Petch, M. Barth, and N. McFarlane) with Leo Donner as the GCSS parameterization expert. It is also planned to invite others onto this group who have expertise in other relevant areas.

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Errata

In the printed issue of IGACtivities No. 33, a co-author was erroneously omitted from the article titled “Wet deposition chemistry in Japan: Key feature of the Japanese Acid Deposition Survey from 1983 to 2002”. Dr. Tsuyoshi Ohizumi of the Acid Deposition and Oxidant Research Center in Japan should have been listed as the third author. We apologize sincerely for this error, which has been corrected on the .pdf version available on-line (<http://www.igac.noaa.gov/newsletter>).

~ Sarah Doherty, IGACtivities Editor

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