Proposal for IGAC Task
ITCT-Lagrangian-2k4

One goal of the Intercontinental Transport and Chemical Transformation (ITCT) program is to understand the chemical transformation and removal processes of aerosols, oxidants and their precursors during the intercontinental transport process. To make this understanding possible, measurements from a Lagrangian platform would be ideal; i.e., a platform that moves with an air mass during the total transport process. Such an ideal is not possible due to the limited range and endurance of existing aircraft. A practical approximation to this ideal, is a "psuedo-Lagrangian" study, where one or more aircraft make multiple, sequential sampling flights into the same air mass during the time required for the intercontinental transport of the air mass. Such a pseudo-Lagrangian study constitutes the IGAC Task described here.

In summer 2004 a large international group of scientists will conduct a field program in the North Atlantic region. The program will focus on the study of emissions of aerosol and ozone precursors over North America, their chemical transformations and removal during transport to and over the North Atlantic, and their impact upon Europe. The program will be organized and funded by several agencies including NOAA (U.S.) through the NEAQS – ITCT 2004 program (http://www.al.noaa.gov/2004/), and NASA (U.S.) through the INTEX-NA program (http://cloud1.arc.nasa.gov/intex-na/). The European contribution, Intercontinental Transport of Pollutants (ITOP), includes contributions from a UK university consortium (funded by NERC), DLR (Germany) and CNRS (France). Each of these programs has its own regionally focused goals and deployments, but together they provide coverage from the source regions on North America, through the transport pathways over the North Atlantic, and over the receptor regions of Europe. The IGAC Task proposed here, ITCT-Lagrangian-2K4, is an organizational and analysis effort that will, within the individual project goals, coordinate the disparate programs into a pseudo-Lagrangian framework. That is, we will combine data from the multiple observational platforms collected at different stages during the transit of a polluted air mass across the North Atlantic.

The ultimate goal is the direct observation of the evolution of the aerosols, oxidants and their precursors from emission over North America, trans-Atlantic transformation and transport, and impact on aerosol and oxidant levels over Europe. Within this overall objective there are several distinct sub-objectives:

- Determination of the photochemical oxidant and aerosol formation potentials in polluted air masses originating in North American emission regions and their chemical evolution as they are transported out over the North Atlantic. This will involve quantification of physical and chemical loss processes of emitted species over North America, and detailed characterisation of radical chemistry, VOCs and their degradation properties, the NO_{xy} budget and physical and chemical aerosol properties in the exported air masses. The contribution of natural sources (e.g. wild fires, biogenics, stratosphere, lightning) to oxidant and aerosol distributions will also be investigated.

- Characterisation of the dynamical processes responsible for pollutant transport out of the North American planetary boundary layer (cyclonic air streams, fronts, convection, landsea breezes, orographic effects) and the importance of mixing/layering processes in the long-range transport and chemical transformation of pollutant plumes as they cross the North Atlantic.

- Quantification of the export of North American pollutants to the background atmosphere, their subsequent fate over the North Atlantic and beyond (e.g. over Europe) and possible impact on climate. This encompasses questions such as: are polluted layers producing or destroying ozone? Do these ageing air masses contain significant levels of anthropogenic aerosols and how is their chemical composition changing as they age?

- Determination of the possible import of North American, and possibly Asian or other, pollutants into the boundary layer over Europe where they may be contributing to background levels of pollutants and regional air quality.

The observational platforms and modeling activities currently proposed are quite extensive. In the source region and over the Western North Atlantic, NOAA will operate the WP-3D aircraft (with in situ gas phase and aerosol instrumentation plus radiation measurements), a remote sensing aircraft (with an ozone and aerosol lidar) and the Ronald H. Brown research vessel (with in situ and remote gas-phase and aerosol instrumentation plus radiation measurements). In that same region, NASA will operate the DC-8 and the P3-B, both with in situ and remote gas-phase and aerosol instrumentation. Within the framework of ITOP the BAe-146 will operate over the central Atlantic from the Azores, and the German DLR and French CNRS Falcons will operate over the Eastern Atlantic and Western Europe. These seven aircraft will also be coordinated with as many as five other research aircraft over North America. These include the Canadian MSC Convair 580 and the Cal Tech/ONR/CIRPAS Twin Otter conducting detailed aerosol and cloud microphysical studies, the NSF/Harvard/COBRA program (operating the Wyoming King Air) conducting a carbon budget study, the U.S. Dept. of Energy G-1, and the University of Maryland Aztec aircraft. These (possibly) twelve aircraft will be coordinated with a variety of ground sites including the AIRMAP network in New Hampshire, Harvard Forest in Massachusetts, a site on the Gulf of Maine, several other U.S. state networks, the PICO-NARE site on the Azores, and the German ATMOFAST lidar and high altitude surface sites.

It is hoped that programs making measurements on commercial aircraft (i.e. MOZAIC, CARIBIC) will also participate in this task by making data available for the period of the measurement intensive. In the case of MOZAIC at least 1-2 flights are made daily across the Atlantic between European cities and, for example, New York or Boston collecting high temporal resolution data on O_3, CO and NO_y. CARIBIC makes less frequent flights (1-2 per month) but with a more comprehensive instrument package that includes measurements of NO_x, non-methane hydrocarbons, halogenated species and aerosols.

Data from current and planned satellite platforms will contribute to the post-campaign analyses, and in some cases will be used for flight planning. Table 1 lists the satellite data sets that provide measurements of tropospheric species and biomass fire information. In addition, the near-real time visible and infrared imagery from GOES and METEOSAT and the winds derived from this imagery will provide flight-planning guidance. Satellite data that have been particularly useful for flight planning include aerosol products from TOMS, SEAWIFS, and

MODIS, O_3 products from TOMS, and the fire data from MODIS. In 2004 we plan to utilize GOME and SCIAMACHY NO_2 columns and perhaps MOPPIT CO columns as well.

Table 1. Tropospheric chemistry and aerosol data sets

Instrument	Species	Vertical Resolution in Troposphere	Current Status
TOMS	O_3, H_2O, Aerosol	Column	Operational since 1979
GOME	O_3, NO_2, CH_2O, SO_2	Column	Operating, but problems with data transmission
MOPPIT	CO	Column & 2 levels	Injured, but operating
SEAWIFS	Aerosol	Column	Operational since 1997
MODIS	Aerosol, fire hot spots	Column	Near real time data available
MISR	Aerosol	Column	On Terra with MODIS
BIRD	Fire hot spots	------	Launched 2001
SCIAMACHY	O_3, CO, NO_2, CH_2O, SO_2, Aerosol	Column	Near real time data available
MIPAS	O_3, H_2O, CO	Limb data in UT	Operational on Envisat
AURA-TES	O_3, H_2O, CO, HNO_3, SO_2	2-4 km resolution	Scheduled to be launched in early 2004
CALIPSO	Aerosol, Clouds	100 m	2004 launch scheduled

Many theoretical groups will be involved in forecasting for flight planning and in modeling for interpretation of results. These include global models (e.g. Harvard, NOAA/U, Iowa, U. Cambridge, MPI-Mainz, CNRS), regional models (U. New Hampshire, Environment Canada) and Lagrangian trajectory models (NOAA, U. Cambridge/Leeds, CNRS).

Coordination of a program to synthesize results from a pseudo-Lagrangian experiment over the North Atlantic requires an international research framework appropriate for IGAC; that coordination is central to the task proposed here.

1. Research Plan. The organization and analysis outlined above comprises four steps: review of previous results, instrument comparison activities (to ensure that measurements on the disparate platforms can be accurately integrated without confounding measurement uncertainties), flight coordination during the field deployment, and post-deployment analysis. ITCT-Lagrangian-2K4 will establish a steering group (SG) to oversee pre-campaign planning, development of a coordinated flight strategy and post-campaign data analysis. The SG will include the task coordinators, representatives from the participating aircraft and forecasting and modeling groups, and other interested scientific participants (see section 6 for suggested SG members).

1.1 Retrospective Analysis of Previous Pseudo-Lagrangian Results. During the NARE 1993 and NARE 1997 studies aircraft were operated on both the North American and European sides of the Atlantic. It was not the primary aim of these experiments to perform a Lagrangian study.

These missions were more exploratory in nature, but events may have occurred during both studies when anthropogenically influenced air masses were sampled before and after transport across the Atlantic. However, examination of the data sets for such cases has been rather limited. The first effort of the ITCT-Lagrangian-2K4 task will be to coordinate more extensive analysis. There are two periods to be examined. First, during NARE 1993 the NCAR King Air, operated in the Gulf of Maine by NOAA, characterized an air mass leaving the U.S. Trajectory calculations suggest the British C-130 may have sampled this same air mass during its return flight from Halifax to Britain. Second, during NARE 1997 the NOAA WP-3D characterized air masses off the U.S. east coast. In one case the British C-130 flying from the Azores may have sampled a characterized air mass after substantial transport. In a second case the DLR Falcon flying over Europe may have sampled one of these air masses over Europe. Analysis of other datasets (e.g. MOZAIC, lidar, aircraft) may also reveal interesting insights into the transport pathways and chemical signatures of pollutant plumes transported across the North Atlantic.
The analysis of these earlier data has two primary goals. First, is to see if measured levels of relatively inert tracers, such as carbon monoxide and long-lived NMHCs, support the indications from trajectory calculations that the same air mass was sampled. Second, is to see if indications of processing can be discerned in the measured levels of more reactive species such as shorter-lived NMHCs, oxides of nitrogen, ozone, and other oxidants. The success and problems identified in these analyses of the early studies will serve as a valuable guide to the field implementation during 2004.

1.2 Instrument Comparison Activities. For the pseudo-Lagrangian approach to be successful, it is essential that the aircraft involved make measurements that are equivalent within quantified uncertainties. Quantifying measurement uncertainty establishes an objective, defensible basis upon which the pseudo-Lagrangian analysis can be based. In effect, a unified observation system is created. Comparison exercises will take place before, during, and after the 2004 field mission. The three general phases envisioned for the comparisons are outlined below, followed by a proposed strategy for their implementation.

Evaluation of standards (pre, during, post-mission): Comparison of compressed gas standards should be performed at least once for the different in-situ gas phase instruments on the participating platforms for NO, CO, CO_2, SO_2 and volatile organic compounds (VOCs). Comparison of ozone standards should also be performed. Effort will also be made to evaluate instrumental sensitivities to HNO_3 between the aircraft and shipborne instrumentation by sampling from a characterized permeation device. IC standards will be exchanged between investigators utilizing aerosol composition measurement systems such as PILS.
Direct comparison of measurements (pre, during, post-mission): Prior to field deployment, running instruments in the lab or in the field side-by-side is an excellent way to test performance. This is more easily done for some instruments than others, so this will be up to the various investigators to arrange as desired. During the mission joint flights of two aircraft, overflights of the ship, Ronald H. Brown, and overflights of ground sites will provide data from which instrument performance may be critically assessed. Such overflights provide an opportunity to compare a large variety of gas-phase, aerosol-phase, meteorological, and radiative parameters.
Indirect comparison of measurements (during and post-mission) While sampling in close physical proximity provides useful information, other opportunities exist to evaluate instrument performance by examining data taken during normal flight procedures. For example, the

CO/CO_2 ratio should be approximately conserved for some time during transport over water, suggesting that aircraft and ship data in an urban plume might be usefully compared in this regard. Further, free tropospheric ozone levels should be comparable between the in situ and remote measurements if taken within a well-defined volume and relative short time of one another. These comparisons-of-opportunity can provide useful additional data with which to evaluate instrumental performance between the various participating platforms

Proposed Strategy:

- *Species*: With regard to the pseudo-Lagrangian analysis, the measurements of most importance to compare include the important gas-phase oxidant and aerosol precursor and tracer species: CO, CO_2, NOx, NOy, O_3, SO_2 and VOCs including oxygenates (it may be useful to specify a finite list of VOCs on which to concentrate effort.) For aerosols, likely comparisons include size resolved number density, and chemical composition as measured by PILS and aerosol mass spectrometer systems. Although perhaps secondary to the pseudo-Lagrangian analysis, the opportunity will be taken to compare the measurements of other important species as well, including the HOx family (OH, HO_2, and RO_2), peroxides and carbonaceous aerosol.
- *Organization:* A small group, with one person from each major organizational group (NASA, NOAA, etc), to devise a strategy, with input from all, act as referees, and attend to logistical details for comparisons. IGAC may be able to play an important role in this organization.
- *Formality:* Semiformal. Each data set will be reduced and submitted independently to the referee, accompanied by the estimated uncertainties, before assessing the comparison data. When all data of a chemical for a comparison flight are submitted, they are released to all study participants. Referees encourage comparison participants to look for non-recoverable errors.
- *Develop comparison matrix* - wingtip-to-wingtip, aircraft to ground or ship, US to European - so that each platform is tied into the comparison through at least one comparison study. Ideally, each comparison occurs at least twice, and is done over a range of important parameters (i.e., altitude, water vapor, possibly interfering pollutants.)

1.3 Flight Coordination. The pseudo-Lagrangian approach to trans-Atlantic transport involves three arenas of flight operations: over the source and outflow region of North America, over the mid-Atlantic and over the inflow region of Europe. Flight coordination will differ in these different regions. On the North American side of the Atlantic, ITCT-Lagrangian-2K4 will not attempt to formulate flight plans or coordinate flights of the various aircraft. Each of the aircraft will have a variety of program goals to meet and will conduct flights according to those goals. However, these goals are such that they ensure that air masses with strong anthropogenic influence leaving North America will be well characterized by measurements during multiple aircraft flights as well as measurements from other platforms. The responsibility of ITCT-Lagrangian-2K4 will be to closely monitor the flights that are made in the source region, to monitor forecast trajectories for these air masses, and to alert aircraft in the central and eastern Atlantic of possible interception opportunities. On the European side, efforts will be coordinated under the ITOP umbrella. A coordinated strategy for flight planning will be developed in collaboration with North American participants.

ITCT-Lagrangian 2K4 will coordinate the development of planning tools and procedures for identifying potential events that aircraft over the mid-Atlantic and Europe can intercept. As part of this planning process, further analysis of trajectories and tracer calculations over periods of several years will be needed in order to establish the most suitable locations for the mid-Atlantic and eastern Atlantic flights. The strategy for coordinating flights must be discussed and developed during planning for the 2004 study.

1.4 Post-Campaign Analysis. A combination of data analysis and modeling will be used to address the scientific objectives of individual programs and also the wider objectives of ITCT-Lagrangian-2K4. The SG will communicate with the separate science teams to assure that the Lagrangian related data sets are available to all of the science teams involved, and will coordinate the analysis of the data from the pseudo-Lagrangian point of view. Other data such as those collected on commercial aircraft, by sondes/lidar, by satellites may also become part of the ITCT-Lagrangian-2K4analysis data set. The SG will organize joint workshops to discuss the results and to identify possible papers to describe key findings. Finally the SG will deal with such issues as publicity arising from any new findings in consultation with the IGAC office.

2. Natural Variability. This project will cover a very limited time window – approximately 6 weeks in summer. The choice of season was made for several reasons: over North America it is the time of maximum photochemical production of oxidants and aerosols, of maximum biogenic emissions of hydrocarbons that provide much of the fuel for this photochemistry, of enhanced stagnation episodes that allow the primary emissions, oxidants and aerosols to collect over the source regions, and a time of consistent northeastward transport of continental air masses to the North Atlantic troposphere. This narrow time window necessarily limits the investigation of natural variability on time scales longer than weeks. Contrasts with previous NARE studies, which were conducted in the early spring, late summer and early fall, will provide some information on seasonal variability.

However, in the interpretation of the results, it is necessary to consider the possibility of significant inter-annual variability. Importantly, the largest source of year-to-year variability of winter weather in the extra-tropical Northern Hemisphere is the North Atlantic Oscillation. Its influence on the variability of transport in the summer over the North Atlantic is relatively weak, but still significant. To assess inter-annual variability, longer-term datasets, such as those from ozone sonde networks, AERONET and programs on commercial aircraft (MOZAIC, CARIBIC) can be used together with analysis of model results. Analysis of satellite data will also make an important contribution to our understanding of inter-annual variability of long-range transport of pollutants over the North Atlantic region.

3. Timetable of Research Activities. (At this point the timetable is still tentative for the process after the field deployment. The combined science teams will finalize the timeline.)

- The planning for the study began in early 2002 and is still ongoing.
- Review of previous results will be completed before field deployment.
- Planning for coordinated field activities (tools, modeling, flight strategy etc.)
- Field deployment will be conducted in July and August, 2004.
- Workshop(s) to discuss results and outline possible manuscripts.

- Manuscripts will be submitted for publication within 18 months after the completion of the field deployment.
- The completion date for the task will be the publication of the special journal section(s) describing the results of the study.

4. Quality Assurance and Data Plan. Quality assurance is discussed in detail in Section 1.2 above. The schedule for the Data Plan is: (At this point the timetable is still tentative for the process after the field deployment. The combined science teams will finalize the timeline.)

- A "preliminary" data archive will be created during the field deployment. This will gradually evolve into the "final" data archive as the data reduction process is completed.
- A "final" data archive with merging of data sets on different time bases will be created 9 months after the completion of the field deployment.
- A data workshop will be held within 12 months of the completion of the field deployment.
- The "final" data archive will be released to the public domain 6 months after the workshop.

5. Plans for peer-reviewed manuscripts. As the final product, we intend to submit at least one, and probably several, papers to the special journal section that is developed for the larger NEAQS-ITCT-ITOP-INTEX study. The specific journal will be chosen by consensus of the various science teams. The paper(s) will synthesize the results of the pseudo-Lagrangian analysis of the photochemical processing and removal of oxidants, aerosols, and their precursors during the trans-Atlantic transport of air masses containing North American emissions.

6. Coordination

a. Task Coordinators.

David Parrish
NOAA Aeronomy Lab
325 Broadway R/AL7
Boulder, CO 80305-3328, USA
(+1-303) 497-5274
(+1-303) 497-5126 (fax)
e-mail: parrish@al.noaa.gov

Kathy Law (IPSL/CNRS)
Service Aéronomie Boite 102,
Université Pierre et Marie Curie,
Tour 15 Couloir 15-14, 4 Place Jussieu,
75252 Paris Cedex 05, France
00-33-1-4427-3865
00-33-1-4427-3776 (fax)
e-mail: kathy.law@aero.jussieu.fr

b. Steering Group

Daniel Jacob (NASA INTEX)
Claire Reeves (UK ITOP)
Hans Schlager (DLR)
Valerie Thouret (MOZAIC)
Andreas Stohl (NOAA NEAQS-ITCT)
Plus Task Coordinators

7. Plan for educational and capacity building efforts. This task seeks to perform organizational and analysis efforts that will coordinate disparate, pre-existing programs. As such it does not have educational and capacity building efforts separate from those of the component programs. However, each of these components has its own efforts. For example, the PICO-NARE program is a joint project between the Universidade dos Açores and Michigan Technological University. The interactions will expand the educational capacity of both institutions.