INTERCONTINENTAL TRANSPORT AND CHEMICAL TRANSFORMATION (ITCT)

A Research Activity within the International Global Atmospheric Chemistry Program (IGAC)

WHITE PAPER

I. Introduction

The intercontinental transport of photochemical pollution currently is attracting considerable interest. There are increasing indications that chemical pollutants, even compounds with reasonably short lifetimes, can be detected at great distances from their sources. The interest in the problem is further heightened by questions regarding how long-range transport may change with climate variability.

Previous studies carried out within the framework of the International Global Atmospheric Chemistry Project (IGAC) have documented that photochemical pollution can be transported into the remote atmosphere. However, no attempts have been made to systematically consider the complexities of chemical transformations over intercontinental scales or to quantify, beyond reporting isolated events, the amounts of these short-lived compounds that are transported over these distances. The challenge of doing this is great. The short-lived pollutants are highly variable in space and time, even in the remote atmosphere. Field measurements must investigate this variability in sufficient detail so that the controlling chemical and meteorological processes are correctly identified and quantified. Monitoring must determine trends and identify sources. Models must correctly capture the variability and decipher the trends.

In order to effect this systematic study, a coordinated international research program is required. The goal of this program is to provide a better understanding of the intercontinental transport and chemical transformation (ITCT) of anthropogenic pollution and to assess the consequences of this pollution. The aim of this white paper is to present the historical context of the issues, to indicate how the required research fits into the IGAC framework, and to outline a plan for development of an IGAC research activity.

II. Historical perspective: *Regional issues and evidence for regional scale transport*

The global dissemination of anthropogenic pollution has been a focus of environmental concern for over two generations. Over the past decade, this concern has centered on the emission of long-lived, radiatively important trace species and the influence that their increased concentrations may have on global and regional climate. However, the chemical properties of anthropogenic pollution are also recognized as a global environment problem.

Over two generations ago, the long-range transport of chemical pollution became a concern with the recognition that lead, principally associated with the use of tetraethyl lead as a motor fuel additive, was being distributed globally. This finding coupled with the known toxic properties of lead caused its removal from fuels as well as cessation of other applications. Likewise, the observation of the long-range transport of chemical pesticides, such as DDT (dichloro-diphenyl-trichloroethane) and other organochlorine compounds, and their ecological damage, has been a subject of study for many decades and continues to the present.

The present interest focuses on species that have an atmospheric lifetime of days to months. Substances with the longer lifetimes are reasonably uniformly distributed on a hemisphere-wide basis. Those with the shorter lifetimes can be significantly redistributed from their sources across national boundaries and into marine regions that border the continents or under favorable conditions over even longer distances. There is evidence that such transport is involved in acid rain and in the anthropogenically perturbed levels of ozone, aerosols, and certain long-lived volatile organic compounds (VOCs).

Potential adverse effects associated with acidic deposition became a concern in the 1970s in North America and Europe. This deposition was causing acidification of lakes and there was concern that there was also potential for damage to forests. The increase in acid deposition was linked to emissions from industry, electric power plants and automobiles. The oxides of sulfur (SO_x) and oxides of nitrogen (NO_x) that were emitted from the sources were further oxidized in the atmosphere to sulfuric and nitric acid and subsequently incorporated into aerosols and clouds. These acids and their precursors could be transported hundreds or, perhaps, thousands of kilometers from their sources, crossing national boundaries. This was recognized as an international problem in North America and Europe.

Ozone, which is formed by photochemical reactions involving NO_x and volatile organic compounds (VOCs), was long recognized as one of the principal constituents of urban

photochemical smog. However, in the 1980s, ozone pollution was identified as a regional problem and the transport of ozone across national boundaries was viewed as an international issue in North America, Europe and, most recently, in Asia.

Like ozone, atmospheric aerosols are generated by chemical processes in the atmosphere involving gas-phase precursors from anthropogenic and natural sources. They may also be generated mechanically (wind, cultivation, etc.) and chemically (industrial combustion, biomass burning, etc.). The intercontinental transport of wind-borne dust has been observed for many years. Satellites have followed the plumes of smoke from forest fires over thousands of kilometers. The radiative properties of aerosols have become the focus of the atmospheric science community associated with the effect that these particles can have on the global climate. Interest in the optical properties of aerosols also is related to the influence these particles have on visibility. Visibility affects air transportation and detracts from the esthetics of scenic features. Most recently, findings have related fine particle concentrations to health effects. This has stimulated considerable new interest in the sources and processes that control the atmospheric aerosol distribution.

The broad range of effects associated with the chemical properties of the atmosphere has generated a major international program aimed at studying and understanding the processes that control those properties.

III. Inception of ITCT

The International Global Atmospheric Chemistry Project (IGAC) was established in response to the growing concern about the chemical changes in the global atmosphere, which are described above, and their potential impacts on humanity. The aims of IGAC are to:

- Develop a fundamental understanding of the processes that determine atmospheric composition.
- Understand the interactions between atmospheric chemical composition and biospheric and climatic processes.
- Predict the impact of natural and anthropogenic forcing of the chemical composition of the atmosphere.

At the inception of IGAC, it was recognized that the composition and chemistry of the atmosphere are dependent on climatic, ecological, geophysical and anthropogenic variables. A

systematic study of the atmosphere must be able to account for the complex interactions among all these variables. However, over the globe these variables are strongly dependent on location. For these reasons, five major regions were defined as research foci: marine, tropical, polar, boreal and mid-latitude regions. Two of the activities established under the IGAC marine research focus were the North Atlantic Regional Experiment (NARE) and the East Asian/North Pacific Regional Experiments (APARE). The aim of these two research foci was to determine the influence that the industrial regions of continents that rim the North Atlantic and North Pacific have on the chemical composition of those marine atmospheres. Since its inception, NOAA has provided the leadership for the NARE activity.

Recently, IGAC re-focused its structure into three main subject areas: biosphere-atmosphere exchange, photochemistry, and atmospheric aerosols. In addition, IGAC has moved away from regional concepts toward a more global perspective. In this context, it was natural to coordinate the two IGAC regional activities concerned with photochemistry, NARE and APARE, within a single framework that has a global dimension (ITCT) and both gas-phase and particulate-phase science objectives.

The focus of ITCT is to investigate intercontinental transport of anthropogenic pollution and to determine the chemical transformations that occur during this transport. The investigation will be initially focused in the Northern Hemisphere that contains most of the world landmasses, where most of the world's population resides, and where most of the anthropogenic pollution is generated. Four central research questions define the thrust of ITCT:

- **What are the export fluxes of anthropogenic pollutants from the northern mid-latitude continents (North America, Europe, Asia) to the global atmosphere?** Answering this question is critical for assessing the potential of anthropogenic emissions at northern mid-latitudes to affect global atmospheric chemistry. Emission inventories for major pollutants are generally available but need to be compared with atmospheric observations of export fluxes from the large-scale continental regions. These comparisons must be done in a way that accounts for changing emissions and for complications from chemistry and deposition taking place within the continental source regions.
- **What is the ultimate fate of northern mid-latitude pollutants exported to the global atmosphere?** This fate largely determines the global environmental implications of the pollutants. Long-range transport of pollution at northern mid-latitudes by the prevailing

westerlies may affect surface air quality in continents downwind. Deposition to the oceans or to the Arctic may have important ecological implications, while transport to the tropics may be more important from a climatic standpoint. The task here is to better understand the dynamical and chemical evolution of polluted continental air masses in the global atmosphere.

- **How does intercontinental transport of pollution at northern mid-latitudes affect surface air quality?** Import of pollutants from continents upwind may compromise efforts of individual countries to reach air quality goals through domestic emission controls. The importance of international transport of pollution within a continent has long been recognized in this context. The question posed by ITCT is whether intercontinental transport of northern mid-latitude pollution needs to be accounted for as well. Quantifying both the chemical outflow and the chemical inflow for these large-scale source/receptor regions effectively frames the problem of constructing chemical budgets for these regions.
- **What are the implications for global atmospheric chemistry of rapid population growth and industrialization in the tropics?** The tropics will play a growing role in the future for the global budgets of anthropogenic pollutants. One needs to develop the tools to accurately track changing emission inventories, to quantify pollutant export fluxes from major tropical source regions, and to understand the underlying mechanisms. Compared to northern mid-latitudes one may expect large and changing differences in the types of emissions as well as in the meteorological and photochemical environments that determine the export of pollution.

The stage for addressing these ITCT questions has been set by recent findings of IGAC-related research. Some of the findings within the framework of IGAC and their implications are described below.

IV. Recent evidence for long-range transport of anthropogenic pollution

A. Recent Scientific Findings

During the past decade research accomplished by IGAC and its related international programs have discovered some essential chemistry and transport processes that control the long-

range transport of short-lived chemical compounds. The aim of this section is to discuss the recent scientific findings that form the basis for future investigations.

1. *Ozone from North America dominates ozone distribution in North Atlantic during the summer*

Surface measurements have shown that ozone pollution from North America is easily detectable 1500 km downwind from the North American source region [Parrish et al., 1993]. Subsequently, surface measurements, located 3000 km downwind from the sources established that North American pollution enhances O_3 levels in the central North Atlantic, in the spring [Parrish et al., 1998].

These observations provided the basis for the quantitative assessment of the amount of anthropogenic O_3 transported from North America. Parrish et al. [1993] concluded that in the summer, O_3 transported from North America exceeded natural O_3 from the stratosphere in the lower troposphere over the North Atlantic. Further quantitative refinements of the calculations estimate that the total flux of ozone from North America to the North Atlantic in the summer is on the order of 1.0 to 1.6 Gmol/day [Chin et al., 1994; Berkowitz et al., 1996]. These studies conclusively show that during the summer, the O_3 budget in the lower troposphere over the temperate western North Atlantic is dominated by O_3 photochemically produced from O_3-precursors emitted by North American sources.

2. *Emissions from Asia influence chemical composition over the northwest Pacific*

Investigations were made of the long-range transport of atmospheric trace species over the northwest Pacific Ocean [Hoell et al., 1997]. The results have been used to estimate the magnitude of the human influence on the chemical composition of the atmosphere over the region. The region of maximum outflow lay between approximately 20˚ N and 40˚ N. The delivery of NO_x enriched from the Asian continent to the free troposphere and the subsequent redistribution by large-scale dynamics can influence the ozone production over large areas of the pacific basin [Crawford et al, 1997].

3. *Oxidized sulfur over western Pacific attributed to Asian sources*

Measurements were made of the chemical composition of the atmosphere over the western Pacific. These measurements [Thornton et al., 1997] indicated that in the region throughout the well-mixed troposphere the oxidized sulfur, sulfur dioxide and sulfate in aerosols were largely associated with emissions of sulfur dioxide from sources located on the Asian continent. The principal component of oxidized sulfur was sulfur dioxide. This suggests that cloud and precipitation scavenging was an effective removal mechanism for aerosol sulfate.

4. *Mechanism for the transport of North America pollution to the North Atlantic*

Meteorological measurements show that during the summer, inversions can effectively isolate the marine boundary layer (MBL) from the lower midtroposphere aloft, where the majority of the pollutants are transported in highly stratified layers [Angevine et al., 1996]. The processes that form these layers provide an effective mechanism for the transport of continental pollution into the mid- and upper-troposphere of the North Atlantic. In general, these processes likely play an important role in the transport of continental O_3 and O_3-precursors from the eastern coastal regions of all continents into the marine atmosphere that they border.

5. *The role that fronts play in transporting continental pollution*

The primary direction for transport of North American pollution to the North Atlantic in the summertime is toward the northeast. In general terms, this can be viewed as the result of the prevailing westerly winds developing a southerly component as air masses come under the influence of the clockwise circulation of the Bermuda-Azores high. However, warm sector flow ahead of advancing cold fronts has been identified as the most important process for the transport of pollution from the urbanized U.S. East Coast to the North Atlantic [Merrill and Moody, 1996; Berkowitz et al., 1996]. This mechanism provides a means to rapidly and effectively transport large amounts of relatively short-lived pollution over long distances.

6. *The role that the oxides of nitrogen (NOx) and volatile organic compounds (VOCs) play in the O_3 budget of the North Atlantic troposphere*

In the remote, marine troposphere, the concentrations of carbon monoxide (CO) and methane (CH_4) are adequate to support significant photochemical ozone formation. However,

whether this photochemistry produces rather than destroys ozone is determined by the amount of NO_x available [Fehsenfeld and Liu, 1993]. Hence, the export of pollution-produced NO_x from the continental boundary layer can determine the amount of ozone that is produced in the marine troposphere. Analysis of the correlation of NOy and its component species with CO has demonstrated that only a small fraction of the NO_x emitted in the continental boundary layer is transported to the free troposphere [Parrish et al., 2000b]. Lagrangian trajectory analyses independently show that only a limited amount of NO_x is transported from North America to the North Atlantic [Stohl et al., 2000].

Although there is limited transport of reactive nitrogen from the continent to the marine free troposphere, continental NO_x emissions are believed to still have a significant impact on ozone distribution in the remote marine troposphere. Model results suggest that eventual ozone production in the global troposphere from U.S. emissions is about twice as large as the direct export of ozone from the U.S. boundary layer [Liang et al., 1998; Horowitz et al., 1998; Meijer et al., 2000]. The amount of NO_x delivered to the lower troposphere in the western North Atlantic during summer is sufficient to produce, on average, 1 to 4 ppbv/day of ozone throughout the region [Duderstadt et al., 1998; Parrish et al., 1998]. In contrast, the central North Atlantic (e.g., the Azores) is a region of photochemical ozone destruction [Peterson et al., 1998; Atherton et al., 1996; Parrish et al., 1998].

7. Role of aircraft emissions of NO_x relative to lightning and surface emissions in ozone formation

Recent studies have examined the sources of the oxides of nitrogen in the remote free troposphere over the North Atlantic [Ziereis et al., 2000; Meijer et al., 2000]. The studies indicate that emissions from aircraft and lightning in the free troposphere and transport from the surface play a significant role in determining the NO_x distribution and the consequent formation of ozone in the free troposphere. These sources are found to vary with latitude: aircraft emission being relative more important at high latitudes, lightning at low latitudes.

8. Intercontinental transport of emissions from forest fires

The principal sources of CO in the Northern Hemisphere are fossil fuel combustion, biomass burning, and oxidation of methane and non-methane hydrocarbons. Recent research has

focused on and elucidated the importance of boreal forest fires to northern hemispheric summertime CO background concentrations [Wotawa and Trainer, 2000]. Hemispheric background concentrations of CO declined from late 1980's through the middle of the 1990's. This decline was associated with emission control of CO in the industrial countries of the Northern Hemisphere. This trend has been perturbed since that period by a rapid increase in CO emissions associated with biomass combustion due to wild fires in the boreal regions of North America and Russia. The emission of the NO_x, combustion-produced VOCs, and carbon aerosols may also perturb the atmsopheric chemistry in the Northern Hemisphere. The increase in these forest fire emissions is the direct result of climate variability that has produced drought conditions in the boreal regions during the last ten years.

9. The seasonal variation of the anthropogenic influence on the tropospheric O_3 budget

All of the NARE summertime studies in the western North Atlantic have found a positive correlation between CO and O_3. This positive correlation demonstrates that anthropogenic pollution produces ozone in the summer, as discussed above. However, in the wintertime a negative correlation between CO and O_3 is observed, both at surface sites [Parrish, 1993] and in the free troposphere [Parrish et al., 1998; 2000a]. This negative correlation demonstrates that anthropogenic pollution provides a sink for ozone in the winter. The mechanism of this destruction is consistent with the expected reaction of O_3 with some of the primary pollutants (e.g., NO and unsaturated VOCs). This destruction is expected to occur in all seasons. However, photochemical formation of O_3 more than compensates for the destruction in summer, but not in winter. A simple model based on O_3 correlations with CO demonstrates that the anthropogenic contribution to the O_3 budget over the western North Atlantic is positive in the summer, near zero in the spring, and likely negative in the winter [Parrish et al., 1999].

However, it has not been determined if the overall anthropogenic effect on O_3 in the winter is negative further from the continent. In winter, slow photochemical O_3 production likely occurs over longer transport times and distances in the troposphere. Long-range transport of anthropogenic precursors to low latitudes with more photochemical activity could possibly further enhance O_3 formation in the remote marine troposphere. A combination of these processes may compensate for the initial O_3 destruction. These are important questions to

answer in order to gauge properly the seasonal dependence of the global climate warming potential of anthropogenic O_3.

B. Evidence for impacts

There is ample evidence for the impact of long-range transport of ozone, fine particles, and their precursors. The long-range transport of dust particles from Asia and Africa has been extensively documented [Prospero, 1979; Duce et al, 1980; Talbot et al., 1986; Prospero and Savoie, 1989; Xiao et al., 1998]. Estimations also indicate that there is an increasing trend in the mixing ratio of ozone over the Northern Hemisphere [c.f., Marenco, et al., 1994] that is probably associated with anthropogenic influences on the chemical composition of the atmosphere.

More specifically, several reports have recently appeared in the published literature documenting events that give clear evidence for intercontinental transport of ozone, ozone precursors, and other chemical compounds. In general, these events are limited to the spring and fall season in the Northern Hemisphere when zonal flow from west to east is stronger and chemical processing and vertical mixing in the atmosphere are less.

Measurements made from island locations in the Atlantic and Pacific have recorded events of increased concentrations of ozone and carbon monoxide. Measurements [Jaffe et al, 1998a] made at Shemya, Alaska indicate that in the spring and fall there are periods of enhanced CO and O_3 concentrations that were attributable to long-range transport of pollution from Asia. Enhancements in CO attributable to Asian sources have been observed in Shemya, Guam, Midway, and Hawaii [Jaffe et al, 1998b]. Likewise, enhanced concentrations of ozone and carbon monoxide attributable to anthropogenic sources in North America have been observed in the Azores [Parrish et al., 1998] during the spring.

Measurements made during the spring on the West Coast of the United States have detected Asian influence on the levels of several atmospheric species. In 1985 Parrish et al. [1992] found that levels of O_3, peroxyacetyl nitrate (PAN), nitric acid (HNO_3) and the light alkanes were enhanced during periods when trajectory analysis indicated rapid transport from Asia. Effective lifetimes of O_3 and PAN in the marine troposphere were derived from the correlation of the levels of these compounds with the ratios of the alkanes. In 1997 Jaffe et al. [1999] detected enhanced concentrations of CO and PAN that could be identified as byproducts of emissions from the Asian continent. The enhancements in ozone concentrations during these episodes were

too small to be reliably extracted from the background ozone concentrations. However, model simulations undertaken as part of the study reproduced the observed enhancements in CO and PAN, and indicated that the ozone over the western United States was also enhanced. Other model simulations [Jacob et al., 1999] appear to support this conclusion and, also, indicate that significant enhancements in the average ozone concentration over the Western United States might occur if Asian emissions of ozone precursors are significantly increased.

Elevated concentrations of ozone that are attributed to long-range transport from the boundary layer over North America were observed over Europe [Stohl and Trickl, 1999] during an event that occurred in the spring of 1997. Ozone mixing ratios as high as 100 ppbv were measured in the free troposphere. The high concentrations recorded during this episode were explained by the effective transport without significant dilution of boundary layer air that was carried by an ascending air stream at the leading edge of a trough.

Clearly numerous aspects of long-range transport have been widely investigated. What is missing is a systematic and quantitative understanding of the budgets of the important trace species on a hemispheric to global scale. This is particularly true for the O_3 budget; there is as yet no agreement on the relative importance of natural and anthropogenic sources of ozone to the Northern Hemisphere troposphere. One of the major tasks facing ITCT is to provide this quantitative analysis.

V. Research strategies for ITCT

A. Research tools

3-dimensional models are required to quantitatively answering the ITCT research questions outlined in section 3. They must resolve the coupling between transport, chemistry, and aerosol processes on synoptic scales and finer. Such models already exist but their quality is highly uncertain. They need to be constrained and tested with atmospheric observations to a much more deliberate degree than has been achieved so far. One aspect of the experimental design for ITCT therefore involves a close coupling of atmospheric observations and models, in which the models are driven by assimilated meteorological observations for the period of interest, and the observations are targeted towards testing the key relevant features of the models. As discussed below, observations from ground-based, aircraft, and satellite platforms all have important roles to play in this design, both in testing model features, and in providing a wider

base of exploratory measurements. Ship platforms are not specifically discussed but share some of the advantages and disadvantages of ground-based and aircraft platforms.

Ground-based platforms have two primary strengths. First, they provide the opportunity for field programs that simultaneously measure a broad spectrum of gaseous and aerosol species by research grade, prototype instrumentation. Such field studies expand the observational database that allows the identification and investigation of crucial atmospheric processes. During these studies many species can be measured by two or more techniques to allow for intercomparison of results. Such intercomparison efforts are critical for the development and improvement of measurement techniques. Second, ground-based platforms allow long-term observations at relatively low cost. Measurements can be made for a large number of species under well-calibrated conditions. A disadvantage of ground-based measurements is the lack of spatial information, which is critical for interpreting the long-range transport and chemical evolution of air masses. Lidars and sondes give vertical information but only for a few species (ozone, aerosols, water vapor). The principal roles of ground-based measurements in the ITCT context are 1) to expand the observational data base of simulataneous measurements of a broad spectrum of atmospheric species, 2) to providehigh-quality data on seasonal and interannual temporal trends, 3) to identify correlations between species that may provide important constraints for the models, and 4) to extend in time the information gained from intensive field studies.

Aircraft measurements have the advantage of providing large spatial coverage. High-quality measurements can be made from aircraft for a number of species. By designing the flight plans in the context of chemical model forecasts, the observations can be collected in a manner that provides an optimal test of models. The disadvantages of aircraft observations are limitation in flight hours, so that the data are only snapshots in time, and the limits on instrumentation dictated by aircraft payload considerations. This time limitation can be overcome to some extent with regular observations from commercial aircraft or from chartered small aircraft, but the number of measured species is then even more limited and there is less flexibility in the observational strategy.

Satellite measurements can potentially provide global and continuous observations of tropospheric composition and are in that regard ideally suited for the ITCT problem. However, for the foreseeable future only a few species will be observable from space [Singh and Jacob,

2000], the vertical resolution will be coarse (at most a few pieces of information in the troposphere), and the precision will remain limited. All current and planned satellites are in polar orbit and have a return time of 3 days or more for any given spot on the Earth; thus the data are not truly continuous. Measurements from geostationary orbit would provide truly continous observations on the scales of relevance to ITCT but these measurements are still in a technology demonstration phase.

B. Research agenda

We envision a three-pronged research agenda for ITCT involving (1) intensive field studies aimed at investigation of specific processes, (2) long-term observations to place these processes in a seasonal and interannual context, and (3) assessments directed at policy development.

Intensive field studies harness measurements from a number of platforms, together with supporting 3-D chemical transport models, into a concerted experimental design focused on answering a limited set of well-defined questions. To address the ITCT questions, these field studies must involve one or more research aircraft. Some specific plans are given in section 6. One useful tool to optimize the value of the measurements for testing the models will be to operate the models in forecast mode over the course of the intensive field study. These forecasts will then be available to guide flight planning on a day-to-day basis. Integration of satellite observations into the experimental design may also be helpful to place the limited aircraft observations into a broader spatial context. Developing this synergy between aircraft and satellite observations may call for inclusion of satellite validation flights in the aircraft mission plans.

Long-term observations are essential to address the ITCT questions by extending temporally the information from the intensive field studies into a seasonal and multi-year frameworks. Long-term measurement platforms may include ground-based sites, ships and commercial aircraft, and small chartered aircraft. Satellites have been deployed to produce valuable long-term observations but have some important limitations discussed in section 3. The selection of platforms should be made with careful consideration of 3-D model results in order to provide the best test of the models towards addressing the ITCT questions. Eventually, the long-term measurement program may evolve into a monitoring operation to document changes in

anthropogenic emissions from continental source regions and their global implications for atmospheric chemistry.

Assessment. ITCT will undertake periodic assessment of the implication of its research findings in terms of the effects of projected changes in emissions and other forcing variables (e.g., changes in land use or climate) for intercontinental transport of pollutants. In this activity ITCT hopes for a closer interaction between the policy communities of the large industrial countries. We expect that the policy community will provide guidance concerning the assessments of highest priority. These assessment needs will direct the scientific resources of the program towards the most critical uncertainties to be addressed.

VI. ITCT program components

A. International Global Atmospheric Chemistry (IGAC) Program

IGAC is a Core Project of the International Geosphere-Biosphere Program (*IGBP*). The *IGAC* (http://web.mit.edu/afs/athena.mit.edu/org/i/igac/www/index.html) program is focused on understanding the chemistry of the global atmosphere as it relates, in part, to the assimulative (i.e. oxidizing) capacity of the atmosphere and the impact of atmospheric composition on climate and climate on atmospheric composition. The *IGAC* program has several projects that address key aspects of the chemistry of the globe, as well as crosscutting activities that support all projects (*IGAC*, 1998). The research activities of the *IGAC* program are in various stages of readiness; many are underway now and others are still being planned

B. NASA Global Tropospheric Experiment and Earth Observing System

The NASA contribution to the *IGAC/ITCT* is through the Global Tropospheric Experiment (GTE) and the Earth Observing System (EOS). The GTE program supports aircraft missions aimed at understanding natural and anthropogenic influences on the global composition of the troposphere. The next mission, Transport and Chemical Evolution over the Pacific (TRACE-P; http://www-gte.larc.nasa.gov/gte_fld.htm#TRACE), will take place in March-April and is strongly focused on the first two ITCT questions (section 3). The objectives of TRACE-P are: (1) to better understand and quantify the export of environmentally important gases and aerosols, and their precursors, from the Asian continent; and (2) to better understand the processes controlling the chemical evolution of the Asian outflow over the western Pacific.

TRACE-P will use two NASA aircraft, the DC-8 (ceiling 12 km) and the P-3B (ceiling 7 km) operating out of Yokota Air Force Base (near Tokyo, Japan) and Hong Kong. Four chemical forecast models will operate continuously during the mission to guide the day-to-day flight planning so that the TRACE-P measurements may provide the best possible test of the models. Near-real-time satellite measurements from AVHRR, SEAWIFS, GOME, TOMS, and MOPITT will provide additional information to guide the flight plan and to place the aircraft observations in context.

A future mission under consideration by GTE for summer 2004 is the Intercontinental Chemical Transport Experiment (INTEX). INTEX will focus on North America, analyzing chemical outflow/inflow and investigating the associated continental boundary layer chemistry and ventilation processes. It will continue and expand upon the research conducted as part of NARE. The experimental design involves several aircraft operating over the United States and oceanic outflow/inflow regions from bases in Maine, Wisconsin, and California. It also involves ground-based stations in the United States to provide continuous measurements of surface composition, and satellite measurements to place the limited aircraft measurements in a larger-scale context. Beyond ITCT, the scientific objectives of INTEX are directly relevant to the carbon cycle research community; INTEX is thus expected to benefit from collaborations with other NASA programs and across agencies.

The EOS program at NASA supports satellite missions that are of direct interest to ITCT. The CO measurements from the MOPITT instrument, presently in space aboard the Terra satellite, will provide important information on chemical outflow from industrial continents and intercontinental transport of pollution plumes. The TES Fourier transform spectrometer (FTS) instrument, to be launched aboard the Aura satellite in mid-2003, will provide vertical profiles of both ozone and CO in the troposphere. Terra and Aura are polar orbiters; they give global coverage but with relatively sparse temporal density. The GIFTS instrument, presently being developed as a technology demonstration through the NASA New Millennium Program (NMP), will launch the TES FTS technology into geostationary orbit in 2004 and provide continuous mapping of ozone and CO over large scenes (up to 1/6 of the globe). The technology developed through GIFTS could eventually allow for continuous monitoring of chemical outflow from major source regions.

C. ACE (Aerosol Characterization Experiment) – Asia

IGAC has planned a series of Aerosol Characterization Experiments (ACE) that integrate in-situ measurements, satellite observations, and models to reduce the uncertainty in calculations of the climate forcing due to aerosol particles. ACE-Asia is the fourth in this series of experiments and will consist of three focused components in the 2001-2004 timeframe:

1. In-situ and column integrated measurements at a network of ground stations will quantify the chemical, physical and radiative properties of aerosols in the ACE-Asia study area and assess their spatial and temporal (seasonal and inter-annual) variability (2001-2004).
2. An intensive field study will be used to quantify the spatial and vertical distribution of aerosol properties, the processes controlling their formation, evolution and fate, and the column integrated clear-sky radiative effect of the aerosol (Late March through April, 2001).
3. The effect of clouds on aerosol properties and the effect of aerosols on cloud properties (indirect aerosol effect) will be quantified in focused intensive experiments (Spring 2001 and Spring 2002 or 2003).

ACE-Asia has been divided into these three separate components so that measurement campaigns can be carefully focused to address the goals of each component. This structure acknowledges that the various components are in different stages of scientific readiness and have different instrumental, sampling, meteorological, and logistical needs. Each of the focused components of ACE-Asia will have its own Science and Implementation Plan to elaborate its goals, research plan, needed measurements, platforms and investigators.

The ACE-Asia research program involves scientists from several countries including: Australia, China, Japan, United Kingdom, United States, and South Korea. Support for the U.S. participation has come from several federal agencies: National Oceanic and Atmospheric Administration (NOAA), National Science Foundation (NSF), National Aeronautics and Space Administration (NASA), and Department of Energy (DOE). The research team also includes a large number of university scientists, both from the U.S. and abroad.

The ACE-Asia intensive field study will take place in early spring, 2001 at the same time as TRACE-P. The overall goal of ACE-Asia is to reduce the uncertainty in climate forcing caused by aerosols over Eastern Asia and the Northwest Pacific and to develop a quantitative understanding of the multi-phase gas/aerosol particle/cloud system. ACE-Asia will involve two

years of observations from a surface network, in addition to springtime intensive observations with aircraft and ships in 2000 and 2001. Since the goals of TRACE-P and ACE-Asia are complementary, collaboration has been encouraged while maintaining the integrity and independence of each mission.

D. The Atmospheric Chemistry Project of NOAA's Climate and Global Change Program

The Climate and Global Change (CGC) Program is an initiative within NOAA that coordinates and supports relevant NOAA research. The Atmospheric Chemistry Project of CGC continues to direct its grant support to research that aids projects of the *IGAC* Program, particularly the *ITCT* research program (http://www.al.noaa.gov/WWWHD/pubdocs/ITCT/). NOAA is augmenting the IGAC/ITCT activities by developing capabilities to better quantify the transport of pollution into and from North America. Emissions from North America can impact European air quality. In addition, pollutants transported into the U.S. become part of the "background" that defines a limit for air quality management. The research is focussed on the long-lived pollutants, CO, ozone and fine particles. The research attempts to define the location of the sources of the pollutants and determine the nature of those sources. These sources include natural sources, such as forest fires, volcanoes, and wind-blown dust; manmade sources, such as transportation and heavy industry; and in-route sources, including ships and aircraft. Research conducted to date has focused on modeling studies of impacts from biomass burning and transport from Asia to North America and measurements and modeling of transport and chemistry in the North Atlantic region (NARE). The proposed research will investigate transport across the Pacific and Atlantic Oceans and North Polar Region. In addition, the chemical transformation and deposition that may occur in the oceanic and polar regions will be studied. The program will:

- conduct airborne exploratory studies of airflow from the Pacific into the western U.S. in early 2001,
- develop surface monitoring capabilities and deploy an intensive field study in 2002 to investigate and quantify trans-Pacific transport,
- build on existing modeling efforts to quantify transport of pollution from Asia to North America, and

- analyze existing data and conduct further measurements to determine the influence of biomass burning.

E. European Export of Precursors and Ozone by Long-Range Transport: A Study in EUROTRAC-2 (EXPORT-E2)

The *EXPORT-E2* project is intended to coordinate European activities in *ITCT* and in doing so to raise the profile of this phenomenon with policy makers. It will build on the recent experience of a major aircraft campaign held at DLR Oberpfaffenhofen where three aircraft (UK Meteorological Office C-130, DLR Falcon, and CNRS Mystère) investigated various aspects of the transport of pollution from Europe in well-defined meteorological events (*EXPORT*).

F. BIBLE

The Biomass Burning and Lightning Experiment (BIBLE) Phase C, conducted by the Japanese Earth Observation Research Center (EORC), National Space Development Agency of Japan (NASDA) is scheduled to conduct measurements in the Western Pacific during December 2000 which will provide data supplemental to the TRACE-P mission. The goal of BIBLE is to study tropospheric chemistry (natural and anthropogenic processes) in the tropical Asia/Pacific region. Measurements of ozone, ozone precursors and other photochemical quantities will be made aboard a Gulfstream II aircraft. In addition, BIBLE Phase C will be making measurements of lightning over the Western Pacific during the ferry flights to and from Northern Australia and Indonesia.

G. PEACE

The Pacific Exploration of Asian Continental Emission (PEACE) mission, also conducted by EORC/NASDA, will take place in January 2002 and will also provide data complementary to TRACE-P. Using the Gulfstream-II aircraft mentioned in the previous section, PEACE will make measurements of the seasonal excursion of the continental outflow from Asia.

H. PHOBEA

The recently conducted Photochemical Ozone Budget of the Eastern North Pacific Atmosphere (PHOBEA) aircraft campaign off the northwest coast of the United States in April-May 1999

(http://weber.u.washington.edu/~djaffe/phobea/) revealed layers of high ozone and aerosols transported across the north Pacific from the Asian continent (D. Jaffe is the PHOBEA mission scientist). A second PHOBEA mission conducted concurrently with TRACE-P would be of great value for investigating the long-range transport and chemical evolution of the Asian outflow sampled with the TRACE-P aircraft

I. Pico International Chemical Observatory, a component of the North Atlantic Regional Experiment (PICO-NARE)

A two-year study of atmospheric chemistry in the lower free troposphere (FT) over the Azores is being organized by Richard Honrath of Michigan Technological University (http://www.cee.mtu.edu/~reh/azores/pico). The objective of this project to make direct measurements over the North Atlantic of pollutants transported from North America and. Since most transport occurs in the free troposphere, these measurements must be made above the marine boundary layer (≈ 2 km). In the entire central North Atlantic, there is only one mountain that reaches above this altitude: Pico. We therefore will make measurements from the summit of Pico for a 2-year period, in order to determine the frequency and magnitude of transport events that disperse ozone and carbon monoxide. Additional measurements will be made by collaborating researchers from Portugal and the U.S., to the extent possible within the station's space and power constraints.

The site will be operated by Michigan Technological University and funded by U.S. National Oceanic and Atmospheric Administration (NOAA). Setup of the semi-permanent measurement site on Pico requires the development of research collaborations with, and granting of permissions by, scientists and government officials in the Azores Islands and in mainland Portugal. These collaborations are established and the measurement site on Pico will be initiated in the summer of 2001. It is expected that this site will form the basis for continuing measurements beyond the period the two year period currently funded.

J. Pacific Landfalling Jets Experiment (PACJET)

The *PACJET* study (http://www.etl.noaa.gov/programs/pacjet/) will be carried out in January and February, 2001 to develop and test methods to improve short-term forecasts of damaging weather on the U. S. West Coast in landfalling winter storms emerging from the

meteorological data sparse Pacific Ocean. Instrumentation for measurement of ozone and carbon monoxide will be included to collect an exploratory *ITCT* data set.

References

Angevine, W. M., M. Trainer, S. A. McKeen, and C. M. Berkowitz, Mesoscale meteorology of the New England coast, Gulf of Maine and Nova Scotia: Overview, *Journal of Geophysical Research*, *101*, 28,893-28,901, 1996.

Atherton, C. S., S. Sillman, and J. Walton, Three-dimensional global modeling of the transport and photochemistry over the North Atlantic Ocean, *Journal of Geophysical Research*, *101*, 29,289-29,304, 1996.

Berkowitz, C. M., P. H. Daum, C. W. Spicer, and K. M. Busness, Synoptic patterns associated with the flux of excess ozone to the western North Atlantic, *Journal of Geophysical Research*, *101*, 28,923-28,933, 1996.

Chatfield, R. B., Anomalous HNO_3/NO_x ratio of remote tropospheric air: Conversion of nitric acid to formic acid and NO_x?, *Geophysical Research Letters*, *21*, 2705-2708, 1994.

Chin, M., D. J. Jacob, J. W. Munger, D. D. Parrish, and B. G. Doddridge, Relationship of ozone and carbon monoxide over North America, *Journal of Geophysical Research, 99*, 14,565-14,573, 1994.

Corbett, J. J., and P. S. Fischbeck, Emissions from ships, *Science*, *278*, 823-824, 1997.

Duce, R. A., C. K. Unni, B. J. Ray, J. M. Prospero, and J. T. Merrill, Long-range atmospheric transport of soil dust from Asia to the tropical North Pacific: Temporal variability, *Science*, *209*, 1522-1524, 1980.

Duderstadt, K. A., et al., Photochemical production and loss rates of ozone at Sable Island, Nova Scotia during the North Atlantic Regional Experiment (NARE) 1993 summer intensive, *Journal of Geophysical Research, 103*, 13,531-13,555, 1998.

Fehsenfeld, F. C., and S. C. Liu, Tropospheric ozone: Distribution and sources, in Ozone in the Troposphere, edited by C. N. Hewitt and W. T. Sturges, pp. 169-231, Elsevier Applied Science, New York, 1993.

Horowitz, L. W., J. Liang, G. M. Gardner, and D. J. Jacob, Export of reactive nitrogen from North America during summertime: Sensitivity to hydrocarbon chemistry, *Journal of Geophysical Research, 103*, 13,451-13,476, 1998.

Jacob, D. J., et al., Origin of ozone and NO_X in the tropical troposphere: A photochemical analysis of aircraft observations over the South Atlantic basin, *Journal of Geophysical Research*, *101*, 24,235-24,250, 1996.

Jacob, D. J., J. A. Logan, and P. P. Murti, Effect of rising Asian emissions on surface ozone in the United States, *Geophysical Research Letters*, *26*, 2175-2178, 1999.

Jaffe, D., et al., Transport of Asian air pollution to North America, *Geophysical Research Letters*, *26*, 711-714, 1999.

Jaffe, D., A. Mahura, J. Kelley, J. Atkins, P. C. Novelli, and J. Merrill, Impact of Asian emissions on the remote North Pacific atmosphere: Interpretation of CO data from Shemya, Guam, Midway and Mauna Loa, *Journal of Geophysical Research*, *102*, 28,627-28,635, 1997.

Jaffe, D. A., L. N. Yurganov, E. Pullman, J. Reuter, A. Mahura, and P. C. Novelli, Measurements of CO and O_3 at Shemya, Alaska, *Journal of Geophysical Research*, *103*, 1493-1502, 1998.

Kasibhatla, P., et al., Do emissions from ships have a significant impact on concentrations of nitrogen oxides in the marine boundary layer?, *Geophysical Review Letters*, *27*, 2229-2232, 2000.

Lary, D. J., A. M. Lee, R. Toumi, M. J. Newchurch, M. Pirre, and J. B. Renard, Carbon aerosols and atmospheric photochemistry, *Journal of Geophysical Research*, *102*, 3671-3682, 1997.

Lawrence, M. G. and P. J. Crutzen, Influence of NO_x emissions from ships on tropospheric photochemistry and climate, *Nature*, *402*, 167-170, 1999.

Liang, J., L. W. Horowitz, D. J. Jacob, Y. Wang, A. M. Fiore, J. A. Logan, G. M. Gardner, and J. W. Munger, Seasonal budgets of reactive nitrogen species and ozone over the United States, and export fluxes to the global atmosphere, *Journal of Geophysical Research*, *103*, 13,435-13,450, 1998.

Liu, S. C., et al., A study of the photochemistry and ozone budget during the Mauna Loa Observatory Photochemistry Experiment, *Journal of Geophysical Research*, *97*, 10,463-10,471, 1992.

Marenco, A., H. Gouget, P. Nedelec, and J.-P. Pages, Evidence of a long term increase in tropospheric ozone from Pic du Midi data series - Consequences: Positive radiative forcing, *Journal of Geophysical Research*, *99*, 16,617-16,632, 1994.

Merrill, J. T., and J. L. Moody, Synoptic meteorology and transport during the North Atlantic Regional Experiment (NARE) intensive: Overview, *Journal of Geophysical Research*, *101*, 28,903-28,921, 1996.

Oltmans, S. J., et al., Summer and spring ozone profiles over the North Atlantic from ozonesonde measurements, *Journal of Geophysical Research*, *101*, 29,179-29,200, 1996.

Parrish, D. D., C. J. Hahn, E. J. Williams, R. B. Norton, F. C. Fehsenfeld, H. B. Singh, J. D. Shetter, B. W. Gandrud, and B. A. Ridley, Indications of photochemical histories of Pacific

air masses from measurements of atmospheric trace species at Pt. Arena, California, *Journal of Geophysical Research* 97, 15,883-15,901, 1992.

Parrish, D. D., J. S. Holloway, R. Jakoubek, M. Trainer, T. B. Ryerson, G. Hübler, F. C. Fehsenfeld, J. L. Moody, and O. R. Cooper, Mixing of anthropogenic pollution with stratospheric ozone: A case study from the North Atlantic wintertime troposphere, *Journal of Geophysical Research, submitted*, 2000a.

Parrish, D. D., J. S. Holloway, M. Trainer, P. C. Murphy, G. L. Forbes, and F. C. Fehsenfeld, Export of North American ozone pollution to the North Atlantic Ocean, *Science, 259*, 1436-1439, 1993.

Parrish, D. D., T. B. Ryerson, J. S. Holloway, G. J. Frost, M. Trainer, G. Hübler, and F. C. Fehsenfeld, Removal of reactive nitrogen from polluted air masses transported from the continental boundary layer, *Journal of Geophysical Research, in preparation*, 2000b.

Parrish, D. D., T. B. Ryerson, J. S. Holloway, M. Trainer, and F. C. Fehsenfeld, New directions: Does pollution increase or decrease tropospheric ozone in Winter-Spring?, *Atmospheric Environment, 33*, 5147-5149, 1999.

Parrish, D. D., M. Trainer, J. S. Holloway, J. E. Yee, M. S. Warshawsky, F. C. Fehsenfeld, G. L. Forbes, and J. L. Moody, Relationships between ozone and carbon monoxide at surface sites in the North Atlantic region, *Journal of Geophysical Research, 103*, 13,357-13,376, 1998.

Peterson, M. C., R. E. Honrath, D. D. Parrish, and S. J. Oltmans, Measurements of nitrogen oxides and a simple model of NO_y fate in the remote North Atlantic marine atmosphere, *Journal of Geophysical Research, 103*, 13,489-13,503, 1998.

Prospero, J. M., Mineral and sea-salt aerosol concentrations in various ocean regions, *Journal of Geophysical Research, 84*, 725-731, 1979.

Ryerson, T. B., et al., Emissions lifetimes and ozone formation in power plant plumes, *Journal of Geophysical Research, 103*, 22,569-22,583, 1998.

Singh, H. B. and D. J. Jacob, Future Directions: Satellite observations of tropospheric chemistry, *Atmospheric Environment, 34*, 4399-4401, 2000.

Stohl, A., M. Trainer, D. D. Parrish, T. B. Ryerson, and J. S. Holloway, Export of CO and NO_y from the North American boundary layer during NARE 96 and NARE 97, *Journal of Geophysical Research, submitted*, 2000.

Stohl, A., and T. Trickl, A textbook example of long-range transport: Simultaneous observation of ozone maxima of stratospheric and North American origin in the free troposphere over Europe, *Journal of Geophysical Research, 104*, 30,445-30,462, 1999.

Talbot, R. W., R. C. Harriss, E. V. Browell, G. L. Gregory, E. I. Sebacher, and S. M. Beck, Distribution and geochemistry of aerosols in the tropical North Atlantic troposphere: Relationship to Saharan dust, *Journal of Geophysical Research, 91*, 5173-5182, 1986.

Weber, R. J., P. H. McMurry, R. L. Mauldin III, D. J. Tanner, F. L. Eisele, A. D. Clarke, and V. N. Kapustin, New particle formation in the remote troposphere: A comparison at various sites, *Geophysical Research Letters*, *26*, 307-310, 1999.

Wotawa, G., and M. Trainer, The influence of Canadian forest fires on pollutant concentrations in the United States, *Science*, *288*, 324-328, 2000.

Xiao, H., G. R. Carmichael, J. Durchenwald, D. Thornton, and A. Bandy, Long-range transport of SO_X and dust in East Asia during the PEM B Experiment, *Journal of Geophysical Research*, *102*, 28,589-28,612, 1997.

Appendix 1: The Tokyo Workshop

In order to initiate the ITCT planning process a meeting was held in Tokyo, Japan on March 16-17, 2000 at the Frontier Research Systems for Global Change. The aim of the Workshop was to define the objectives of IGAC/ITCT and to identify relationships between ITCT and other science programs both within and outside IGAC. The convenors for the Tokyo Workshop were Hajime Akimoto of the University of Tokyo and FRSGC, Japan; Fred Fehsenfeld of NOAA, USA; and Stuart Penkett of the University of East Anglia, UK. Participating Countries included: China, France, Germany, India, Japan, Norway, Russia, Taiwan, USA, and UK. The Workshop goals were to:

1. Determine how currently planned research will contribute to a better understanding of ITCT.
2. Discuss how to determine the "natural background" levels of ozone and aerosols and to reliably predict how these levels might change with global climate change. How does stratospheric/tropospheric exchange and emissions from aircraft and ships affect the composition in the remote troposphere?
3. Discuss the effects of global climate change on long-range transport and consider how these effects can be evaluated.
4. Determine if this planned research can be coordinated to better understand ITCT.
5. Identify additional research that can complement programs that are already planned.
6. Identify longer-term research activities that are needed.
7. Discuss how network or satellite measurements can promote better understanding.
8. Discuss the role that instruments aboard commercial aircraft presently play and can play in the future to provide needed information.
9. Discuss what types of integrated field studies are needed. Indicate when (seasonal) and where these studies should be carried out.

The presentations and discussions focused on a review of ongoing and planned research programs and a discussion of the new science that will be required to understand the international transport and chemical transformation of anthropogenic pollution.

A. Agenda and presentations

March 16, 2000
8:40 AM Registration (SEAVANS North Building 1F Meeting room)

9:00 AM Welcome:
Taroh Matsuno, Director-General, FRSGC
Hajime Akimoto, Director of Atmospheric
Composition Research Program, FRSGC

9:15 AM Charge to Workshop:
Stuart Penkett, University of East Anglia
Fred Fehsenfeld, NOAA/Aeronomy Laboratory

PART 1: WHERE WE ARE (Fehsenfeld)
9:30 AM to 12:00 noon (wrapped around a break)
What We Know: Review of Research Accomplishments
Several major studies have been undertaken that are concerned at least in part with long-range transport and chemical transformation.
Short presentations (about 10 minutes each, two to five viewgraphs) that indicate:
What are the major findings (focus on most recent (unpublished) findings)?
Are there perceived gaps, disagreements, or inconstancies in, between, among the findings of various studies?
Presentations by:

Yutaka Kondo	Aircraft observational study
Sham Lal	INDOEX and others
Shaw Liu	Present issues after PEM-West
Oystein Hov	Observations using the EMEP data base.
Stuart Penkett	NARE: recent findings.
David Parrish	NARE: status of issues and uncertainties.
Daniel Jacobs	Results from some recent field programs.
Hanwant Singh	Issues raised by SONEX and POLINAT results.
Reg Newell	Comments regarding evolving understanding of long-range transport that can be derived from recent field studies.
Dan Jaffe	Issues raised by recent measurements.
Tim Bates	ACE I & II: status of issues and uncertainties.
Joe Prospero	Understanding long-range transport of fine particle: results from recent field programs.

10:30 AM to 11:00 AM Coffee /Tea Break

12:00 noon to 12:30 PM Discussion and Comment

12:30 PM to 1:30 PM Lunch

1:30 PM to 2:30 PM
Current Status of Modeling Capability (Akimoto)
A number of modeling concepts has been developed to simulate intercontinental transport and chemical transformation. This discussion will provide for a brief review of current modeling capability.
Short presentations (about 10 minutes each, two to five viewgraphs) that indicate:
How well are the important physical and chemical processes captured in current models?
What needs to be improved?
What do model comparisons show?
Presentations by:

Masaaki Takahashi	GCM/Tropospheric Chemistry Coupling Model
Daniel Jacob	Perspective on the status of modeling capabilities.
Michael Trainer	Comments on modeling capabilities (Levy/Carmichael).
Claire Granier	Perspective on the status of modeling capabilities. (Model Intercomparison exercise IGAC-GIM)
Oystein Hov	Perspective on the status of modeling capabilities. (Comparison of model output with measurements)

2:30 PM to 3:00 PM Discussion and Comment

3:00 PM to 3:30 PM Coffee /Tea Break

3:30 PM to 4:30 PM
Added Value: Networks, Satellites and Commercial Aircraft. (Penkett)
Currently, several strategies are being used to provide routine regional, hemispheric and global monitoring of ozone, fine particles and their precursors. This discussion will provide for a brief review of current and expected capability.
Short presentations (about 10 minutes each, two to five viewgraphs) that indicate:
How can new and emerging satellite observations be used to determine transport of continental emission to the remote marine troposphere?
What role can surface sampling networks play in determination of long-range transport and chemical transformation?
How can satellite and network measurements be augmented by instruments carried aboard commercial aircraft?
Presentations by:

Jacobs/Parrish	Satellite observations (Fishman/Thompson).
Sam Oltmans	Network observations.
John Burrows	Satellites observations.
Stuart Penkett	Mosaic/Caribic

4:30 PM to 5:00 PM Discussion and Comment

5:00 PM to 5:30 PM General Discussion, Wrap-up

6:00 PM to 8:00 PM Reception (restaurant "Star Board" in the SEAVANS building, A-Mall 2F)

March 17, 2000

PART 2: WHAT IS PLANNED (Fehsenfeld)

9:00 AM to 11:30 AM (wrapped around a break)

Planned research and Identification of Important Gaps.

Brief reports of major programs planned to address long-range transport from the continents. (To Europe, to Asia, to North America)

Short presentations (about 10 minutes each, two to five viewgraphs) that indicate:

How will the currently planned research contribute to a better understanding of intercontinental transport and chemical transformation (ITCT)?

Are there limitations or gaps in this approach to that question?

Presentations by:

Nikolai Elanski	Siberian study
Hajime Akimoto	Trans-Eurasian study
Mingxing Wang	Chinese conceptual plan
Yutaka Kondo	Future Japanese plan
Tim Bates	ACE Asia
Daniel Jacob	TRACE Pacific
Reg Newell	Meteorological perspective: can these programs add significant new information concerning ITCT.
Dan Jaffe	Proposed programs.
Granier/Penkett	European programs that will address ITCT.
Oystein Hov	NILU China Network
John Burrows	Eurotrac II Satellite

10:30 AM to 11:00 AM Coffee /Tea Break

PART 3: WHERE WE ARE GOING

11:30 AM to 12:00 PM

I Existing Programs: Closing the Gaps.

This discussion and comment period will address:

Given time constraints, can existing programs be complemented to fill the gaps?

Can this planned research be coordinated to better understand ITCT?

12:00 noon to 1:00 PM Lunch

1:00 PM to 2:00 PM

II Additional Research Needed: What Don't the Continents Put There? (Akimoto, Fehsenfeld, Penkett)

This discussion and comment period will address:

What are the "natural background" levels of ozone and aerosols?

Influence of stratospheric/tropospheric exchange (O_3, CO, H_2O).

Influence of surface exchange (particles, particle precursors, and ozone precursors).

Marine surfaces

Land surfaces

How do emissions from aircraft and ships affect the composition in the remote troposphere?

2:00 PM to 3:00 PM
III. Additional Research Needed. The Effect of Climate Change.
How these effects can be evaluated?
This discussion and comment period will address:
Effect of climate change on long-range transport?
Effect of climate change on the "natural background".

3:00 PM to 3:30 PM Coffee /Tea Break

3:30 PM to 5:00 PM
IV Identifying long-term research activities: perspectives on future research needs.
This discussion and comment period will address:
What types of integrated field studies are needed?
When (seasonal) and where should these studies be carried out?
What additional modeling or measurement tools are needed?
How can monitoring (by networks, satellies and commercial aircraft) contribute to these goals?

5:00 PM to 6:00 PM General Discussion, Wrap-up

A. Conclusion from the Workshop

There was a broad agreement among the participants that:

- A better understanding of the role of intercontinental transport and the intervening chemical transformation is required.
- Regional and seasonal variations in the distribution of long-lived pollutants must be observed and their sources identified.
- Trends in their distribution must be monitored and the causes understood.
- Given the magnitude of the problem, where possible existing programs should seek closer cooperation.
- Future programs should be designed to promote expanded international cooperation.

As a first step, an ITCT Coordination Committee has been established to identify opportunities for cooperative research. This will include the following members (others to be added later):

North America	F.C. Fehsenfeld, D. Parrish, H. Singh, D. Jacob,
Europe	S.A. Penkett, Ø. Hov, J. Burrows, C. Granier, K. Law
Asia	H. Akimoto, Y. Kondo, S. Liu, S. Lal

B. Future meeting and meeting aims

Two planning meetings (*USA/Europe*) have been scheduled for the coming year. The first of these meeting is scheduled for December 2000 and will be held in Boulder, CO, USA. This meeting will be restricted to the ITCT Coordination Committee. The aim of the meeting is to finalize a draft science plan for ITCT that can be circulated to all the participants of the Tokyo workshop. We hope to submit research plan to IGAC and IGBP by 1 May 2001.

The second meeting will be held in Europe in the spring of 2001. The aim of this meeting is to draft a plan for an activity to assess the extent and impact of long-range transport of air pollution. We envision that assessments will be produced on a recurring basis every four to five years. This meeting will involve the ITCT Coordination Committee and selected representatives from the policy communities of the large industrial countries. We expect the policy community will provide guidance concerning the type of information to be contained in the assessment that would be most helpful to that community. We hope to have a draft of the first assessment prepared

Appendix 2. The NOAA Component of ITCT

NOAA will undertake to augment ITCT activities by developing capabilities to better quantify the transport of pollution into and from North America. Emissions from North America can impact European air quality. In addition, pollutants transported into the U.S. become part of the "background" that defines a limit for air quality management. The research will initially focus on the long-lived pollutants, CO, ozone and fine particles. The research will attempt to

define the location of the sources of the pollutants and determine the nature of those sources. These sources will include natural sources, such as forest fires, volcanoes, and wind-blown dust; manmade sources, such as transportation and heavy industry; and in-route sources, including ships and aircraft. The proposed research will investigate transport across the Pacific and Atlantic Oceans and North Polar Region. In addition, the chemical transformation and deposition that may occur in the oceanic and polar regions will be studied. The program will:

- conduct airborne exploratory studies of airflow from the Pacific into the western U.S. in early 2001,
- develop surface monitoring capabilities and deploy an intensive field study in 2002 to investigate and quantify trans-Pacific transport, and
- analyze existing data and conduct further measurements to determine the influence of biomass burning at high latitudes.

A. Exploratory Field Study in 2001

During January and February 2001, the Winter Storm Reconnaissance, 2001 program will operate the NOAA Gulfstream IV SP aircraft from Hawaii. Simultaneously, the PACJET (Pacific Landfalling Jets Experiment) program will operate the NOAA WP-3 Orion from California. The primary goal of both of these field programs is to improve meteorological forecasts for storms that strike the U.S. Pacific Coast. The areas of operation of the two aircraft (mid to eastern Pacific) are within the primary transport pathways of pollution between Asia and North America. In addition, the mid-Pacific is a region of intense stratosphere-troposphere exchange associated with breaking Rossby waves. These programs offer us the opportunity to carry out an exploratory study of the pollutant concentrations and transport in this region.

On a noninterference basis, we will measure ozone from the Gulfstream IV and ozone and carbon monoxide (CO) from the WP-3. The mid-Pacific ozone data from the Gulfstream IV will provide the basis for gauging the influence of stratosphere-troposphere exchange on the tropospheric ozone budget of the Pacific region and, when combined with trajectory analysis, will give an indication of transport of anthropogenic pollution during the winter season. The eastern Pacific ozone and CO data from the WP-3 will indicate the influence of the transport of Asian pollution to the western U.S. coast. It will also gauge the effect of the recirculation of North American emissions, and will track the inflow of natural ozone from the stratosphere-

troposphere exchange in the mid-Pacific. The results of this exploratory study will provide important guidance for planning the much more extensive field mission planned for the following winter-spring period.

B. Development of surface monitoring capabilities

In order to better track the long-range transport of pollution to and from North America and to monitor the change in the pollutant distributions with time, additional surface monitoring sites and upgrades to existing sites are planned.

Azores. Two measurement sites are planned for the Azores in the central North Atlantic Ocean. These projects are outgrowths of the CO, ozone, and nitrogen oxides measurements made at Terceira Island in 1993 [*Peterson et al.*, 1998; *Parrish et al.*, 1998]. The Pico Exploratory Atmospheric Chemistry Observatory (PEACO) is a planned two-year study of atmospheric chemistry in the lower free troposphere (FT) over the Azores. The measurements will be made at the summit of Pico mountain on Pico Island at an altitude of 2200 m, a height that is frequently in the FT according to ozone soundings made at the adjacent island of Terceira [*Oltmans et al.*, 1996]. Despite the significant logistical and organizational difficulties involved in operating on a mountaintop with no existing access or electrical power, measurements are scheduled to begin in early summer 2001. Government permission has been received to set up the site and make measurements for a 2-year period.

The primary objectives of the measurements are to quantify the impact of continental outflow on the budget of ozone over the North Atlantic and to assess the relative importance of ozone and ozone precursor export. However, it is expected that the measurements will be used for a variety of other purposes, including aerosol studies. Initially, the measurements will include CO, ozone, and standard meteorological observations. Future measurement additions include black carbon, nonmethane hydrocarbons, and nitrogen oxides. Space and electrical power are quite limited and instruments must be remotely monitored and controlled.

The Portuguese Meteorological Institute is planning a long-term Global Atmospheric Watch (GAW) site in the marine boundary layer (MBL) in the Azores. The value of an Azores GAW site was mentioned in a recent WMO informal report ("Final Report of the Consultation of Experts on Carbon Monoxide Instrumentation for Remote GAW Sites, Geneva, 8-10 September 1999"). Measurements at this site, in conjunction with the mountaintop measurements, will

allow the investigation of FT/MBL exchange and the budgets of ozone and its precursors in the MBL of the central North Atlantic.

NOAA Baseline Observatories. The NOAA Climate Monitoring and Diagnostics Laboratory (CMDL) operates long-term atmospheric observatories at Pt. Barrow, Alaska and Mauna Loa, Hawaii. Presently, the measurements are aimed at climate forcing and ozone-depleting agents such as CO_2, CH_4, O_3, N_2O, CFC's, aerosols and solar incoming and upwelling radiation. In addition, persistent organic pollutants and mercury have been added recently at Barrow. Measurements at the observatories will be enhanced as part of the ITCT Program. During Spring 2001, a pilot intensive will be held at Mauna Loa (Springtime TRansport of Effluents from Asia to Mauna Loa – STREAM) during which additional measurements such as continuous CO, persistent organic pollutants, mercury, nitrogen oxides, aerosol chemistry, and hydrocarbons (Proton Transfer Mass Spectrometry and GC-MS instruments) will be conducted. The results of this pilot campaign will be used to evaluate possible observatory participation during the proposed 2002 intensive field study.

Northwestern U. S. Pacific Coast. An initial study of the factors that influence the airquality on the western fringes of the United States will be undertaken during the spring of 2002 at sites that will be identified and developed during the coming year (2001). Possible sites include Trinidad Head, CA, where CMDL presently operates both a Dobson ozone spectrophotometer and an ozonesonde station. In addition, the NASA Advanced Global Atmospheric Gases (AGAGE) program measures various chlorofluorocarbons (CFCs) at this site. Both programs operate through cooperation with Humboldt State University. CMDL plans to expand its background measurements of air-quality parameters at this site in the future and to work with other agencies and universities to develop a distributed baseline observatory on the west coast of the U.S.

C. Field Study in 2002

During Spring 2002, NOAA will conduct airborne measurements of the concentrations of ozone, fine particles, their precursors, the photochemical intermediates, and other photochemical products, as well as other atmospheric parameters. The study will be based on the U.S. West Coast, and will investigate how inflow from the Pacific basin affects the chemical processing and

removal of compounds of anthropogenic origin that influence the regional budgets of ozone and fine particles downwind over the continental U.S.

The objectives for this program are to provide information that pertains to:

(1) *Marine boundary layer and free troposphere:*

- inflow to U.S. continent, upwind of coastal shipping lanes to address issues of intercontinental transport [*Parrish et al.,* 1992; *Jaffe et al.,* 1999; *Jacob et al.,* 1999];
- new particle formation, composition, growth, and evolution in low-hydrocarbon environments [*Weber et al.,* 1999];
- influence of coastal ship traffic on marine boundary layer (MBL) NO_x levels, ozone, and aerosols; mixing and oxidation in ship plumes [*Corbett and Fischbeck*, 1997; *Lawrence and Crutzen,* 1999; *Kasibhatla et al.,* 2000];
- horizontal and vertical distributions of ozone, CO, aerosol, etc. during inflow periods, to which U.S. emissions are added. This will provide a benchmark for future investigation of hemispheric impacts of increasing Asian emissions.
- provide data to constrain heterogeneous processing of HNO_3 on aerosols [*Liu et al.,* 1992; *Chatfield*, 1994; *Jacob et al.,* 1996; *Lary et al.,* 1997].

(2) *Continental boundary layer and free troposphere:*

- impact of West Coast emissions on downwind ambient concentrations, in contrast to the East Coast situation studied in previous NARE missions. Ozone production efficiency of NO_x emissions from large combustion sources when added to relative clean marine air;
- vertical transport, oxidation, and sinks of ozone, aerosols, and their precursors emitted from coastal metropolitan areas into marine air masses, through the contrast of upwind (marine) profiles with downwind (impacted continental) profiles;
- ozone and aerosol processing and growth as marine air is advected over pine forests, where pinenes and their oxidation products contribute very significantly to the biogenic volatile organic compounds (BVOC);
- vertical transport, oxidation, and export of anthropogenic NOx, hydrocarbons, PAN, and ozone into the free troposphere (FT); human influence on FT ozone from West Coast inland [*Parrish et al.,* 1993; *Chin et al.,* 1994; *Parrish et al.,* 1998];
- inflow from Central America and Mexico and the impact on U.S. aerosol loading from sources from Central America and Mexico;

- West Coast urban/point source NOx plume oxidation processes, where prevailing non-methane hydrocarbon (NMHC) concentrations are lower and not as strongly dominated by isoprene (sources in deserts, sources in pine forest) in contrast to those in the southeastern US [*Ryerson et al.*, 1998]

(3) how chemical processing on aerosols influences ozone formation;

(4) how the atmospheric oxidation leading to ozone formation also leads to aerosol formation;

(5) how atmospheric chemistry influences the growth and/or the chemical composition of aerosols; and

(6) the effects of dynamic (mixing) factors on the observed concentrations of gas phase and aerosol species.

D. Influence of biomass burning at high latitudes

For many years, the influence of biomass burning on regional air quality and climate variability has been recognized. Much of the emphasis of these studies has been to determine the influence of intentional burning related to agriculture and land use in regions between 20ºN and 20ºS. However, biomass burning at high latitude in the Northern Hemisphere has important regional and even global consequences. In the summer of 1995, elevated concentrations of CO were observed in the southeastern and eastern United States. This CO could not be explained by emissions from known anthropogenic sources in the United States. The source of this CO proved to be a large forest fire that was burning in the boreal forest of northwestern Canada (*Wotawa and Trainer*, 2000). Based on the forest area burned in Canada annually, it is estimated that on-average these fires amount to approximately 17% of the annual U.S. CO emissions.

Beyond regional impacts in the United States, during intense fire years the forest fires in the high latitudes (> 40ºN) may have an influence on the summertime CO background over the Northern Hemisphere. These fires are unintentional, often generated by lightning. They generally occur in the more remote boreal forests. These "wildfires" are modulated by regional climate variability and are most prevalent during periods of decreased precipitation. These fires emit significant quantities of trace gases CO, CO_2, CH_4, VOCs, and NO_x. It is very important that hemispheric and global trends of trace gases can be estimated considering the influence of boreal fires.

Several research projects will be undertaken to better understand the roles played by these fires.

- Long-term CO measurements data will be investigated to search for inter-annual variability consistent with seasonal variations and magnitude of high-latitude fires.
- Determine the influence of NO_X and VOC emissions for high-latitude fires on summertime ozone levels over North America.
- Determine the chemical composition of gases and fine particles emitted by these fires and the chemical evolution of those emissions once they enter the atmosphere.
- Determine the influence of these aerosols on hemispheric climate forcing and variability with particular emphasis on the their impact in the North Polar Region.