THE ICARTT STUDY – LINKING REGIONAL AIR QUALITY TO INTERCONTINENTAL TRANSPORT

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ABSTRACT

There is a growing awareness of the importance of intercontinental transport on the global background of ozone and fine particles. The lowering of regulatory thresholds for these pollutants in the U.S. has farther increased the interest in understanding the impact of intercontinental transport or regional air quality. During the last decade the U.S. National Oceanic and Atmospheric Administration (NOAA) has conducted a series of intensive field campaigns to separately identify the factors responsible for poor air quality on a regional basis and to examine the role of intercontinental transport on global and regional air quality. In the summer of 2004 NOAA will join several groups in North America and Europe that had independently developed plans for field experiments aimed at developing a better understanding of the factors that shape air quality in their respective countries and the remote regions of the North Atlantic. While each of these programs has regionally focused goals and deployments they share many of the same goals and objectives and the proposed study areas overlap significantly. ICARTT (International Consortium for Atmospheric Research on Transport and Transformation) was formed to take advantage of this synergy by planning and executing a series of coordinated experiments to study the emissions of aerosol and ozone precursors their chemical transformations and removal during transport to and over the North Atlantic. The capabilities represented by the consortium (e.g., an extensive surface network, 12 instrumented aircraft, and a research vessel) will allow an unprecedented characterization of the key atmospheric processes. The combined research conducted in the programs that make up ICARTT will focus in three main areas: regional air quality, intercontinental transport, and radiative balance in the atmosphere.

BACKGROUND

By almost any measure, air quality worldwide has improved significantly over the past thirty years. Process improvements, advances in technology, and the development of more effective, emission reduction strategies have led to a lowering of ambient levels for many pollutants in most of the industrialized world. This success is even more impressive when we consider the population and industrial growth that has occurred during that same period. In many parts of the world the focus of air quality management has changed as the most egregious sources of air pollution have been controlled. In particular, air quality managers working to reduce local pollution levels have extended their emission control efforts from local sources to emissions on regional and continental scales. In the United States this broader perspective has been motivated, at least in part, by a lowering of the National Ambient Air Quality Standards for ground level ozone and particulate material. These new standards place a premium on regional, multistate, and possibly international cooperation if effective management strategies are to be developed.

Another factor that has had a significant effect on research related to long-range transport is the recognition of the strong linkages between air quality and climate. Clearly, the distinction between these two research areas is, at least in part, simply a matter of perspective and scale. Many of the chemical and meteorological processes of interest are common to both. Also, intercontinental transport is either the starting point or the end point of regional air quality concerns depending, in the case of the U.S., on whether you are on the west coast (inflow) or east coast (outflow).

One focus of NOAA's climate research has been global-scale transport and transformation processes, which is linked to other U. S. based research and international efforts through the International Global Atmospheric Chemistry Program (IGAC). NOAA organized major field campaigns to study pollutant transport from North America to the North Atlantic under the North Atlantic Regional Experiment (NARE) in 1993, 1996 and 1997. NOAA also co-organized major field campaigns to study regional distributions of aerosol properties and their radiative effects as part of the IGAC Aerosol Characterization Experiments (ACE) in 1995, 1997, and 2001. More recently, the transport of Asian pollution to the U. S. west coast was studied in 2002 under the Intercontinental Transport and Chemical Transformation (ITCT) program.

NOAA's Health of the Atmosphere (HoA) research is focused on the atmospheric science that underlies regional and continental air quality, with the goal of enhancing our ability to predict and monitor future changes, leading to improved scientific input to decision-making. The HoA program is a collaborative effort involving several NOAA laboratories and university scientists. Under this program NOAA joined with other federal agencies, university research groups, and interested parties from the private sector to study factors controlling the formation and distribution of ozone and fine particles in a number of settings including: Nashville, TN (1994, 1995, 1999), Atlanta, GA (1999), and Houston TX (2000). The most recent effort was a pilot study in New England in the summer of 2002 – the New England Air Quality Study (NEAQS).

Thus, in recognition of the strong linkage between air quality and climate NOAA will conduct a joint regional air quality and climate change study in the summer of 2004. The study will combine the elements of the previous ITCT and NEAQS studies as part of the 2004 International Consortium for Atmospheric Research on Transport and Transformation (ICARTT). The study will focus on air quality along the Eastern Seaboard and transport of North American emissions into the North Atlantic.

PREVIOUS NOAA RESEARCH ON LONG RANGE TRANSPORT

NARE - The North Atlantic Regional Experiment (NARE) was established by the International Global Atmospheric Chemistry (IGAC) Program to study the chemical processes that occur in the remote marine environment of the North Atlantic. The NARE science objectives were:

- To assess the long range transport of photochemically active compounds and/or their products and determine the impact of this transport on hemispheric air quality.

- To ascertain the effect of these compounds on the oxidative properties and radiation balance of the atmosphere.
- To estimate the amounts of these compounds that are deposited in this marine environment in order to better quantify the potential impact of this deposition on surface sea-water chemistry and marine biological processes.

The initial objective of NARE was to investigate the chemical and transport processes that shape the distribution of ozone over the North Atlantic and to estimate the impact of human-influenced emissions from North America and Europe on the production of tropospheric ozone in this region. Initial research was conducted in 1991 with measurements of outflow of North American pollution at surface sites in the Maritime Provinces of Canada [1]. The first major intensive field study was conducted in the summer of 1993. The results were published in two special sections of JGR [101, D22 and 102, D11].

Two additional field intensives were conducted in early spring, 1996 and late summer/early fall, 1997, which were reviewed in the IGACtivities Newsletter, Issue No. 24. The interpretation of these studies has focused on the chemical evolution, removal and transport patterns of anthropogenic emissions over the North Atlantic [2,3,4,5,6]

ITCT - 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 East Asian/North Pacific Regional Experiments (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?
- What is the ultimate fate of northern mid-latitude pollutants exported to the global atmosphere?
- How does intercontinental transport of pollution at northern mid-latitudes affect surface air quality?
- What are the implications for global atmospheric chemistry of rapid population growth and industrialization in the tropics?

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.

NEAQS - During the summer of 2002 NOAA deployed one of its research vessels, ***RONALD H. BROWN***, to the Gulf of Maine. The ship was instrumented with a full array of instrumentation to quantify gaseous and aerosol pollution and important meteorological parameters. The ship was used to study the chemical evolution of the New York city and Boston plumes and to track pollution being transport into the Gulf of Maine to coastal New England, and to the North Atlantic. The primary goal of this pilot study was to better understand the dynamics of the marine boundary layer and chemical transport during overwater transport.

FINDINGS RELEVANT TO INTERCONTINENTAL TRANSPORT

These three studies have provided important new insights into magnitude and process that control intercontinental transport. Examples include:

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 [7].

2. A persistently stable marine boundary layer promotes transport of North America pollution to the North Atlantic – Meteorological measurements show that during the summer, the lower atmosphere offshore is frequently strongly stratified. Therefore, pollutants are efficiently transported in discrete and persistent layers [8]. The processes that form these layers provide an effective mechanism for the transport of continental pollution into the lower and mid-troposphere of the North Atlantic.

3. Fronts play an important role in transporting continental pollution to the North Atlantic - 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 [9,10]. This mechanism provides a means to rapidly and effectively transport large amounts of relatively short-lived pollution over long distances.

4. Availability of oxides of nitrogen (NOx) limits O_3 production in 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 [11]. 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 [12].

Lagrangian trajectory analyses independently show that only a limited amount of NO_x is transported from North America to the North Atlantic [13].

5. *Trajectory calculations indicate that there is rapid and efficient transport from Asia to the west coast of the U.S. along frontal systems* [14,15]. - Transport distances over the north Pacific are sufficiently long that Asian airmasses typically encounter multiple weather systems prior to arrival over North America. The compounds showed remarkably little dispersion during transport. However the process of entraining these pollutants into the continental boundary layer does result in strong dilution, which serves to decrease the impact of Asian pollution on the air quality of the U.S. west coast [16].

6. *The chemical mix of airmass transported from Asia to the U.S. west coast can be modified significantly during transport as a result of several mechanisms:*

- Residual NO_x, SO_2, and VOCs react during transport to produce additional ozone and fine particles. PAN decomposition appears to play a significant role in ozone formation [17].
- Stratospheric intrusions and lightening combine to produce additional ozone during transport [15].
- Interaction with clouds effectively deplete soluble pollutants.

7. *The pollution plume leaving the east coast of the U.S. rivals those from Asia in terms of aerosol loading.* International field campaigns during the past 7 years have studied aerosol loading and properties downwind of North America (1996 - TARFOX), Europe (1997 - ACE-2), SE Asia (1999 - INDOEX), and Asia (2001 - ACE-Asia). ACE-2 also used multiple aircraft to measure the first indirect effect of aerosols. More recently, shipboard measurements during NEAQS 2002 along the U.S. East Coast showed that the U.S. pollution plume can be as intense (in terms of aerosol mass concentration, aerosol optical depth, and ozone mixing ratio) as those downwind of India and Asia [18]. It is clear from NEAQS 2002 that particulate organic matter is a large fraction of the total aerosol off the coast of North America compared to the aerosol downwind of India (INDOEX) and Asia (ACE-Asia) [18].

ICARTT

Several groups in North America and Europe have independently developed plans for field experiments in the summer of 2004, aimed at developing a better understanding of the factors that shape air quality in their respective countries and the remote regions of the North Atlantic. For example, NASA and NOAA are planning experiments under the Intercontinental Chemical Transport Experiment - North America (INTEX-NA) and the New England Air Quality Study - Intercontinental Transport and Chemical Transformation (NEAQS - ITCT) 2004 programs respectively while the Europeans (U.K., Germany, and France) are organizing coordinated studies under Intercontinental Transport of Pollution (ITOP). While each of these programs has regionally focused goals and deployments they share many of the same goals and objectives and the proposed study areas overlap significantly. ICARTT was formed to take advantage of this synergy by planning and executing a series of coordinated experiments to study the emissions of aerosol and ozone precursors their chemical transformations and removal during transport to and over the North Atlantic. The capabilities represented by the consortium will allow an unprecedented characterization of the key atmospheric processes. The combined research conducted in the programs that make up ICARTT will focus in three main areas: regional air quality, intercontinental transport, and radiation balance in the atmosphere.

Regional Air Quality

Coordinated measurements made by the ICARTT consortium during the 2004 study can help to understand how emissions from local and distant source regions interact to determine the air quality in northeastern United States and the Maritime Provinces of Canada. The mosaic of ozone and aerosol precursor emissions that influences the air quality over a broad region of northeastern North America provides the opportunity to investigate the chemical evolution of individual source categories and their synergism. The coordinated activities possible through the ICARTT consortium will allow two important undertakings that represent breakthroughs in air quality research.

Intercontinental Transport

One of the goals of the 2004 study is to better understand the chemical transformation and removal processes of aerosols, oxidants and their precursors during intercontinental transport. This information forms the basis for understanding the impact of anthropogenic emissions on regional and global climate and air quality. In order to provide the information to make this understanding possible, it is necessary is systematically study an air mass as it moves from continent to continent. The capability to make this ground-breaking study possible will be available for the first time in 2004.

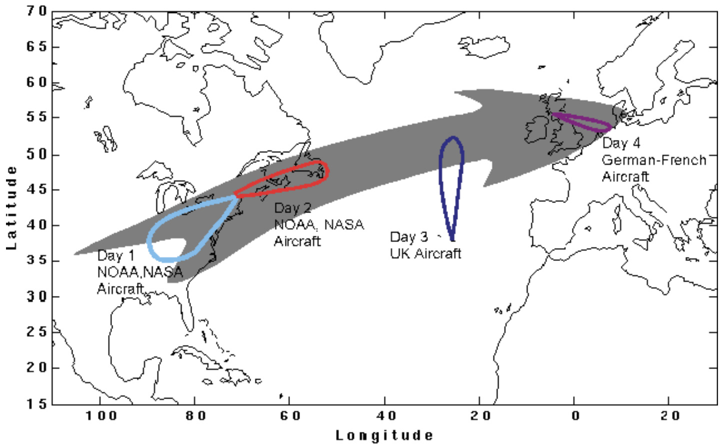

Figure 1. Schematic diagram showing the path of a polluted airmass advected from North America to Europe (gray arrow) and the proposed flight tracks for the NOAA, British and German aircraft.

Radiation Balance

Atmospheric aerosol particles affect the Earth's radiative balance directly by scattering and absorbing sunlight and indirectly by acting as cloud condensation nuclei (CCN), thereby influencing the albedo (first indirect effect, [19]), life-time [20], precipitation [21] and extent [22] of clouds. Aerosol concentrations and their radiative impacts are particularly high in regions downwind of sources where diurnally-averaged clear sky surface forcings range up to 30 Wm-2 [22]. International field campaigns during the past 7 years have studied aerosol properties and their direct radiative effects downwind of North America (1996 - TARFOX), Europe (1997 - ACE-2), SE Asia (1999 - INDOEX), and Asia (2001 - ACE-Asia). During these 7 years of major field programs our scientific tools for measuring aerosols and their radiative properties and our understanding of the complex chemistry and transport processes have evolved tremendously. We propose to use these new tools to quantify the aerosol properties, controlling processes and radiative effects over Northeastern North America and off the U.S. East Coast during ICARTT 2004. Measurements during NEAQS 2002 showed that the U.S. pollution plume can be as intense (in terms of aerosol mass concentration, aerosol optical depth, and ozone mixing ratio) as those downwind of India and Asia (Quinn and Bates, 2003). NEAQS 2002, however, did not specifically measure the direct or indirect radiative effects of this aerosol pollution plume.

PLATFORMS AND PARTICIPANTS

To address these complex and interrelated issues the ICARTT consortium will deploy a comprehensive array of observing systems stretching from North America to Europe. These systems include:

- A coordinated, daily release of ozonesondes.
- A wind profiler network
- Coordinated chemical measurements at surface sites in the U.S., Canada, and Europe
- Thirteen research aircraft will operate in North America and Europe
- The NOAA Research Vessel Ronald H. Brown will operate in the Gulf of Maine to coast of Nova Scotia.
- Instrumented balloons to track air mass movement and pollutant levels.
- Coordinated Satellite overflights

More information on the ICARTT science planning and sampling platforms visit the ICARTT web site at http://www.al.noaa.gov/ICARTT

REFERENCES

1. 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.

2. Cooper, O.R., J.L. Moody, D.D. Parrish, M. Trainer, J.S. Holloway, G. Hübler, F.C. Fehsenfeld, and A. Stohl, Trace gas composition of mid-latitude cyclones over the western North Atlantic Ocean: A seasonal comparison of O_3 and CO, *Journal of Geophysical Research*, *107* (D7), 10.1029/2001JD000902, 2002.

3. Cooper, O.R., J.L. Moody, D.D. Parrish, M. Trainer, T.B. Ryerson, J.S. Holloway, G. Hübler, F.C. Fehsenfeld, and M.J. Evans, Trace gas composition of mid-latitude cyclones over the western North Atlantic Ocean: A conceptual model, *Journal of Geophysical Research*, *107* (D7), 10.1029/2001JD000901, 2002.

4. Cooper, O.R., J.L. Moody, D.D. Parrish, M. Trainer, T.B. Ryerson, J.S. Holloway, G. Hübler, F.C. Fehsenfeld, S.J. Oltmans, and M.J. Evans, Trace gas signatures of the airstreams within North Atlantic cyclones: Case studies from the North Atlantic Regional Experiment (NARE '97) aircraft intensive, *Journal of Geophysical Research*, *106* (D6), 5437-5456, 2001.

5. Stohl, A., M. Trainer, T. Ryerson, J. Holloway, and D. Parrish, Export of NO_y from the North American boundary layer during NARE 96 and NARE 97, *J. Geophys. Res.*, *107*, 10.1029/2001JD000519, 2002.

6. Li, Q., D.J. Jacob, I. Bey, P.I. Palmer, D. B.N., B.D. Field, R.V. Martin, A.M. Fiore, R.M. Yantosca, D.D. Parrish, P.G. Simmonds, and S.J. Oltmans, Transatlantic transport of pollution and its effects on surface ozone in Europe and North American, *Journal of Geophysical Research*, *107* (D13), 2002.

7. 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, *J. Geophys. Res., 103*, pp. 13,357-13,376, 1998.

8. 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.

9. 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.

10. 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.

11. 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.

12. 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*, 2000.

13. 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.

14. Cooper, O. R., C. Forster, D. Parrish, M. Trainer, E. Dunlea, T. B. Ryerson, G. Hübler, F. Fehsenfeld, D. Nicks, J. Holloway, J. Nowak, C. Brock, J. de Gouw, C. Warneke, J. Roberts,F. Flocke J. Moody, A case study of trans-Pacific warm conveyor belt transport: The influence of merging airstreams on trace gas import to North America, *J. Geophys. Res.*, *109*, 2003JD003624, 2004.

15. Cooper, O. R., C. Forster, D. Parrish, E. Dunlea, G. Hübler, F. Fehsenfeld, J. Holloway, S. Oltmans, B. Johnson, A. Wimmers, and L. Horowitz, On the life-cycle of a stratospheric intrusion and its dispersion into polluted warm conveyor belts, *J. Geophys. Res.*, *109*, 2003JD004006, 2004.

16. Hudman, R.C., D. J. Jacob, O. C. Cooper, M. J. Evans, C. L. Heald, R. J. Park, F. Fehsenfeld, F. Flocke, J. Jolloway, G. Hubler, K. Kita, M. Koike, Y. Kondo, A. Newman, J. Nowak, S. Oltmans, D. Parrish, J. M. Roberts, T. Ryerson, ozone production in transpacific Asian pollution plumes and implications for ozone air quality in California, *Journal of Geophysical Research*, submitted, 2004.

17. Brock, C. A., P. K. Hudson, E. R. Lovejoy, A. Sullivan, J. B. Nowak, L. G. Huey, O. R. Cooper, D. J. Cziczo, J. de Gouw, F. C. Fehsenfeld, J. S. Holloway, G. Hübler, B. G. Lafleur, J. A. Neuman, D. K. Nicks, Jr., D. A. Orsini, D. D. Parrish, T. B. Ryerson, D. J. Tanner, M. Trainer, C. Warneke, R. J. Weber, and J. C. Wilson, Chemical and microphysical characteristics of aerosols in the free troposphere near the west coast of North America, *J. Geophys. Res.*, *in-press*, 2004.

18. Quinn, P.K. and T.S. Bates, North American, Asian, and Indian haze: Similar regional impacts on climate?, *Geophys. Res. Let.*, 30, 1555 (2003).

19. Twomey, S., Aerosols, Clouds and Radiation, *Atmos. Environ.*, *25A* (11), 2435-2442, 1991.

20. Albrecht, B.A., Aerosols, Cloud Microphysics, and Fractional Cloudiness, *Science*, *245*, 1227-1230, 1989.

21. Rosenfeld, D., Suppression of rain and snow by urban and industrial air pollution, *Science*, *287*, 1793-1796, 2000.

22. Ramanathan, V., P.J. Crutzen, J.T. Kiehl, and D. Rosenfeld, Aerosols, climate, and the hydrological cycle, *Science*, *294*, 2119-2124, 2001.