International Consortium for Atmospheric Research on Transport and Transformation (ICARTT): North America to Europe—Overview of the 2004 summer field study

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[1] In the summer of 2004 several separate field programs intensively studied the photochemical, heterogeneous chemical and radiative environment of the troposphere over North America, the North Atlantic Ocean, and western Europe. Previous studies have indicated that the transport of continental emissions, particularly from North America, influences the concentrations of trace species in the troposphere over the North Atlantic and Europe. An international team of scientists, representing over 100 laboratories, collaborated under the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) umbrella to coordinate the separate field programs in order to maximize the resulting advances in our understanding of regional air quality, the transport, chemical transformation and removal of aerosols, ozone, and their precursors during intercontinental transport, and the radiation balance of the troposphere. Participants utilized nine aircraft, one research vessel, several ground-based sites in North America and the Azores, a network of aerosol-ozone lidars in Europe, satellites, balloon borne sondes, and routine commercial aircraft measurements. In this special section, the results from a major fraction of those platforms are presented. This overview is aimed at providing operational and logistical information for those platforms, summarizing the principal findings and conclusions that have been drawn from the results, and directing readers to specific papers for further details.

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1. Introduction

[2] Until recently research programs in global climate change and regional air quality have been conducted as separate, albeit related, activities. The investigation of intercontinental-scale transport and chemical transformation processes and radiation balance in the atmosphere have been the focus of the former, while the latter has been focused on the atmospheric science that underlies urban, regional and continental air quality. Clearly, the distinction between the research objectives of these two programs 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

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both the starting point and the end point of regional air quality concerns since any particular region contributes outflow to and receives inflow from that transport.

[3] In recognition of this strong linkage, a joint regional air quality and climate change study, which is described herein, was planned and carried out in the summer of 2004. The study focused on air quality in the eastern United States, transport of North American emissions into the North Atlantic, and the influences that this transport has on regional and intercontinental air quality and climate, with a particular focus on western Europe.

[4] The topics addressed in the present study have a long history. There have been at least three decades of studies aimed, at least in part, at determining the causes of poor air quality outside of urban areas along the east coast of the United States and the transport of polluted air from North America out into the North Atlantic. Some very early studies have been followed by intensive field campaigns conducted along the eastern coast of North America and into the western North Atlantic. Similarly, intensive field programs along the western coast of Europe and the eastern North Atlantic have investigated the impact of polluted air flowing into Europe. To place the planning that preceded the current study into perspective, section 2 provides a brief review of related previous research.

[5] Several independent field studies, each focused on some aspect of climate change and air quality issues over North America, the Atlantic and Europe, were planned for the summer of 2004. Early in the planning it became evident that coordination between these studies would provide a more effective approach to addressing these issues. The International Consortium for Atmospheric Research on Transport and Transformation (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, and their impact downwind on the European continent.

[6] The combined research conducted in the programs that make up ICARTT focused on three main areas: regional air quality, intercontinental transport, and radiation balance in the atmosphere. Although each of the programs had regionally focused goals and deployments, they shared many of the overall ICARTT goals and objectives. The aims and objectives of the individual components that compose the ICARTT program are briefly described in section 3. The capabilities represented by the consortium allowed an unprecedented characterization of the key atmospheric processes. The scope of the study is indicated by the measurement platforms and ground site locations that were operated during the study and are described in section 4. This section also provides general information that can be referenced in publications that describe results obtained from the study and its interpretation.

[7] The goal of this special journal section is to report many of the ICARTT results; sections 5 and 6 highlight some of the particularly important findings. The NASA Intercontinental Chemical Transport Experiment–North America (INTEX-A) and the CO_2 Budget and Rectification Airborne study (COBRA) participated in ICARTT, but will publish their results elsewhere, the former in a separate special section in *Journal of Geophysical Research*.

2. Review of Previous Research Related to ICARTT

[8] The planning for ICARTT was guided by the findings of many studies of regional air quality, long-range pollutant transport and atmospheric radiative forcing that were carried out over the past three decades in the ICARTT research area. Table 1 and the discussion below briefly summarize these studies and give relevant references providing additional information. The lessons learned from previous studies of long-range transport over different parts of the world such as many of the NASA Global Tropospheric Experiment campaigns [*McNeal et al.*, 1998] were also valuable for the planning for ICARTT. The results of all of these studies provide the context for the analysis and interpretation of the ICARTT results.

[9] A series of studies (NACEMS, AMODES, NARSTO-NE-OPS) carried out in the eastern United States and Canada focused on providing the measurements needed for the evaluation of air quality models. The results from these studies indicated that a three-dimensional regional-scale picture of the atmosphere is required to understand and predict local air pollution events. An ongoing program of atmospheric research carried out at Harvard Forest, a rural site near Petersham, Massachusetts provides a chemical climatology for all seasons over several years, which are particularly relevant to the ICARTT study.

[10] Several programs have measured the atmospheric composition of the North Atlantic region. *Zeller et al.* [1977], *Kelleher and Feder* [1978], and *Spicer* [1982] found evidence for the transport of plumes along the eastern seaboard of the United States and out over 100 km or more of the North Atlantic. Measurements in central Nova Scotia, Canada, observed the long-range transport of plumes from urban and industrial sources in the United States, a distance of over 500 km [*Brice et al.*, 1988; *Beattie and Whepdale*, 1989]. The GCE/CASE/WATOX study investigated transport and deposition of aerosols. The NARE program of IGAC studied the effect of long-range transport of chemical compounds on the oxidative properties and radiation balance of the troposphere over the North Atlantic. The SONEX/POLINAT-2 studies focused on the impact of aircraft emissions on the photochemistry in the upper troposphere/“lowermost” stratosphere. The Atmospheric-Ocean Chemistry Experiment (AEROCE) conducted a systematic study of the influence of anthropogenic emissions on ozone and aerosols at island sites in the North Atlantic. *Winkler* [1988] summarized the ozone measurements from 32 research vessel cruises through the Atlantic Ocean. A special journal section (*Journal of Geophysical Research*, *95*(D12), 1990) reported the results of the cruise of the German research vessel Polarstern in 1987. Several NASA sponsored programs identified transport of pollution from North America to the western North Atlantic: *Anderson et al.* [1993] concluded that anthropogenic pollution has a major impact on the budgets of ozone

Table 1. Atmospheric Composition and Radiative Forcing Studies Previously Conducted in the ICARTT Study Region

Study	Dates	References
	Northeastern North American Continental Studies	
North American Cooperative Network of Enhanced Measurement Sites (NACEMS)	summer-fall 1988	*Trainer et al.* [1993] and *Parrish et al.* [1993]
Acid Model Operational Diagnostic Evaluation Study (AMODES)	summer-fall 1988	*Tremmel et al.* [1993, 1994]
North East Oxidant and Particle Study (NARSTO-NE-OPS)	summers 1998, 1999, 2001	*Zhang et al.* [1998] and *Seaman and Michelson* [2000]
Harvard Forest	ongoing, long-term	*Munger et al.* [1998] and *Goldstein et al.* [1998]
	Western North Atlantic Studies	
Global Change Expedition/Coordinated Air-Sea Experiment/Western Atlantic Ocean Experiment (GCE/CASE/WATOX)	summer 1988	special section in *Global Biogeochemical Cycles*, *4*, 1990
Atmospheric-Ocean Chemistry Experiment (AEROCE)	1988 to present	*Prospero* [2001]
North Atlantic Regional Experiment (NARE)	1993–1997	special section in *Journal of Geophysical Research*, *101*(D22), 1996; special section in *Journal of Geophysical Research, 103*(D11), 1998; and *Li et al.* [2002]
SASS (Subsonic Assessment) Ozone and *NOx* Experiment (SONEX) Pollution from Aircraft Emissions in the North Atlantic Flight Corridor (POLINAT-2)	fall 1997	*Singh et al.* [1999] and *Thompson et al.* [2000b]
New England Air Quality Study (NEAQS)	summer 2002	*Bates et al.* [2005]
	Western Europe and Eastern North Atlantic Studies	
Atmospheric Chemistry Studies in the Oceanic Environment (ACSOE)	spring and summer 1997	*Reeves et al.* [2002]
Maximum Oxidation rates in the free troposphere and Testing Atmospheric Chemistry in Anticyclones (MAXOX/TACIA)	summers in late 1990s	*Reeves et al.* [2002]
Atmospheric Chemistry and Transport of Ozone/European Export of Precursors and Ozone by Long-Range Transport (ACTO/EXPORT)	May and August 2000	*Methven et al.* [2003] and *Purvis et al.* [2003]
Convective Transport of Trace Gases into the Middle and Upper Troposphere over Europe: Budget and Impact on Chemistry (CONTRACE)	May and November 2001	*Huntrieser et al.* [2005]
EUROTRAC-TOR	1996–2002	*Schultz et al.* [1997]
Free Tropospheric Experiments (FREETEX)	1996 and 1998	*Carpenter et al.* [2000]
	Aerosol and Radiative Forcing Studies	
Atlantic Stratocumulus Transition Experiment/Marine Aerosol and Gas Exchange (ASTEX/MAGE)	June 1992	special section in *Journal of Geophysical Research*, *101*(D2), 1996
Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX)	summer 1996	special section in *Journal of Geophysical Research*, *104*(D2), 1999, and special section in *Journal of Geophysical Research*, *105*(D8), 2000
Second Aerosol Characterization Experiment (ACE-2)	summer 1997	special issue in *Tellus, Series B, 52*(2), 2000
AEROSOLS99	winter 1999	special section in *Journal of Geophysical Research*, *106*(D18), 2001

Table 2a. Mobile Platforms Involved in the ICARTT Study

	Program, Agency	Emphasis
Aircraft		
Douglas DC8	INTEX NA, NASA	regional distribution of chemically active compounds over North America and their sources (emphasis on free troposphere); outflow from North America
Lockheed WP-3D	NEAQS/ITCT, NOAA	emissions and chemical processing downwind from urban areas and industrial point sources in the northeastern United States (emphasis on boundary layer); outflow from North America
Grumman Gulfstream I	DOE	emissions and chemical processing downwind from urban areas and industrial point sources (emphasis on boundary layer)
Douglas DC-3	NEAQS/ITCT, NOAA	emissions and chemical processing downwind from urban areas and industrial point sources (emphasis on boundary layer)
FAAM BAE 146–301	ITOP, NERC	observations of chemical processing occurring in air masses transported from North America to Europe
Dassault Falcon	ITOP, DLR	measurements in pollution plumes transported from North America including forest fire plumes originating from Canada and Alaska and quasi Lagrangian studies; measurements of emissions from shipping in the English Channel; satellite validation and measurement comparisons
BAe Jet Stream J-31	INTEX NA, NASA	aerosol, water vapor, cloud, and ocean surface radiative properties and effects; satellite validation; regional-scale understanding of anthropogenic aerosol and radiative impacts
Twin Otter	CIRPAS, NSF	the relationship between cloud properties and the properties of the aerosols that are influencing the cloud formation
Convair 580	MSC	the relationship between cloud properties and the properties of the aerosols that are influencing the cloud formation
Ship		
Ronald H. Brown	NEAQS/ITCT, NOAA	chemical composition and aerosol physical and optical properties in the marine boundary layer; emission from ships; long-path radiation-aerosol measurements

and aerosols in the near continent region; and *Fishman et al.* [1990, 1991] identified a strong, summertime ozone maximum extending downwind from North America into the North Atlantic. The NEAQS project deployed the research vessel *Ronald H. Brown* to study the chemical evolution of gaseous and aerosol pollution in the New York City and Boston urban plumes over the Gulf of Maine.

[11] A series of airborne studies have been carried out over the eastern North Atlantic Ocean and western Europe to investigate the impact of transatlantic transport of anthropogenic emissions. ACSOE, which formed the European component of NARE, and MAXOX/TACIA investigated the chemistry and transport of pollutants through aircraft observations from the Azores and the UK, respectively. ACTO/EXPORT investigated both the inflow of anthropogenic pollutants from North Atlantic regions and the uplift and export of European emissions from the surface. The CONTRACE field experiment intercepted several pollutant plumes from North America over Europe.

[12] Several ground-based studies in Europe have investigated long-range transport of pollution arriving in Europe. The influence of intercontinental transport was observed at mountaintop sites during the EUROTRAC-TOR, FREETEX and CONTRACE experiments. Long-range transport events have sometimes been observed at the Mace Head sea level site [*Derwent and Jenkin*, 1991]. This site is located on the western coast of Ireland, where extensive, continuing atmospheric measurements were initiated in

Table 2b. Ground Sites Involved in the ICARTT Study

Location of Site(s)	Program, Agency	Emphasis
Five sites located in northeastern United States	AIRMAP, NOAA	long-term measurement to document and study persistent air pollutants such as O_3 and fine particles in the region
Pinnacle State Park in Addison, New York	ASRC, NOAA, NSF	measurements of aerosol composition, gaseous aerosol precursors, ozone, and solar radiation
11 site radar wind profiler network	NEAQS/ITCT, NOAA, DOE	regional-scale trajectories and transport of air masses
Chebogue Point, Nova Scotia, Canada	NEAQS/ITCT, NOAA, NSF	determination of the frequency and intensity of pollution events crossing the Canadian maritime provinces; study of aerosol processing
12 Station Ozonesonde Network	IONS, INTEX NA, NASA	estimation of the North American ozone budget by profiling ozone from sites across the continent
Pico mountain, Pico Island, Azores, Portugal	PICO-NARE, NOAA, NSF	determination of the composition of the lower free troposphere in the central North Atlantic region
European Lidar Networks	ITOP	identification of atmospheric layers for the surface to 5 km that were influenced by long-range transport not of European origin

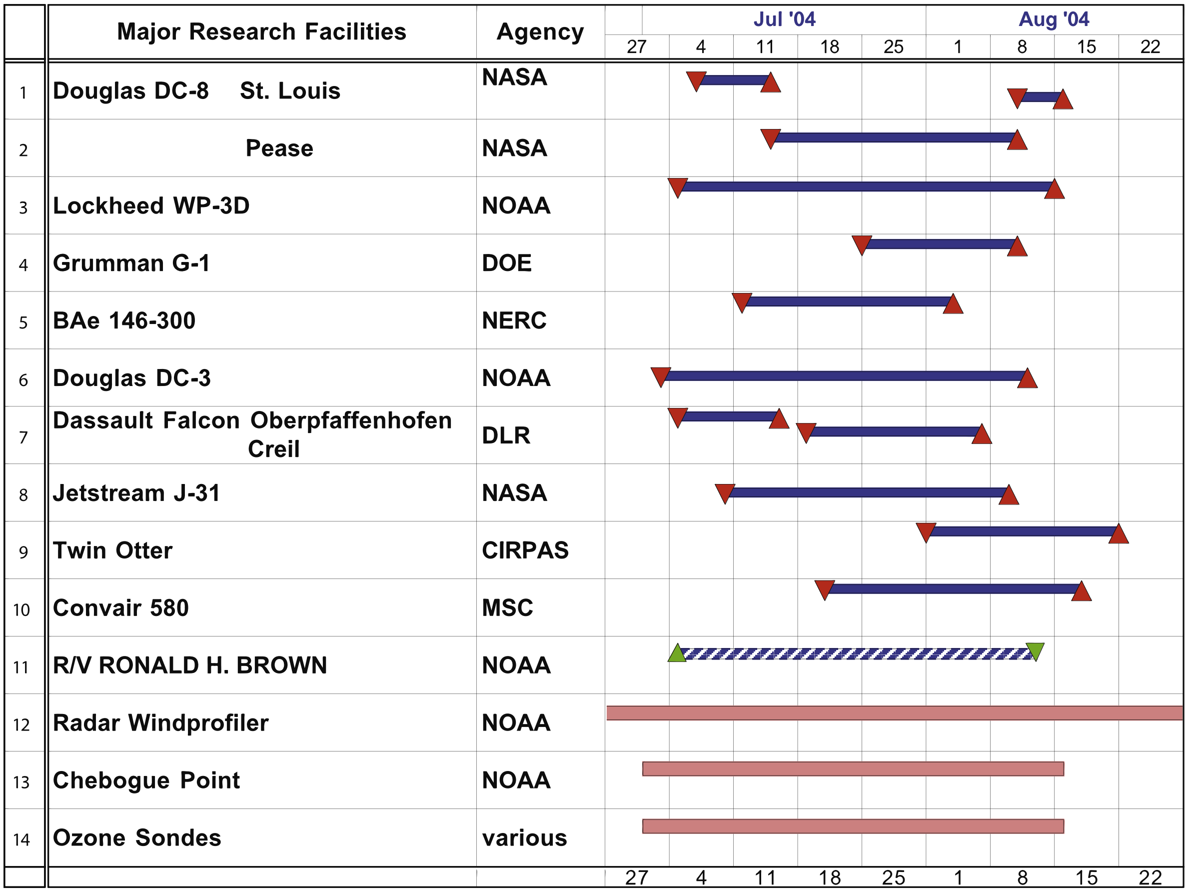

Figure 1. Dates of deployment of the major ICARTT platforms and surface sites.

1987. Data from a network of European LIDARs combined with trajectory analysis have demonstrated several examples of long-range transport of polluted air masses from North America to Europe [e.g., *Stohl and Trickl*, 1999].

[13] The European community has been active in utilizing commercial aircraft for extensive measurements in the free troposphere. The data sets are particularly concentrated over western Europe, the North Atlantic and eastern North America. These programs include Measurements of Nitrogen Oxides and Ozone Along Air Routes (NOXAR) [*Brunner et al.*, 2001], Measurement of Ozone, Water Vapour, Carbon Monoxide and Nitrogen Oxides by In-service Commercial Aircraft (MOZAIC) [*Marenco et al.*, 1998] and Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container (CARIBIC) [*Zahn et al.*, 2004].

[14] Several field campaigns have focused on aerosol transport and transformation over the North Atlantic and the radiative forcing of those aerosols. ASTEX/MAGE investigated the formation and transformation of marine aerosols. TARFOX focused on the direct radiative impacts of aerosols, as well as the chemical, physical, and optical properties, of the aerosols carried over the western Atlantic Ocean from North America. ACE-2 contrasted the aerosol characteristics, processes and effects over the anthropogenically modified North Atlantic with those observed during ACE-1, which was conducted in the minimally polluted Southern Ocean. The shipboard AEROSOLS99 study crossed the Atlantic Ocean from Norfolk, Virginia, to Cape Town, South Africa, and determined the chemical, physical, and optical properties of the marine boundary layer aerosol.

3. Components of ICARTT

[15] During the summer of 2004, ICARTT coordinated the activities of several independently planned research programs. The coordinated programs involved extensive measurements made from aircraft, a research vessel, and several ground stations located in the northeastern United States, Nova Scotia, the Azores, and western Europe. Tables 2a and 2b list the principal measurement platforms and ground stations, the programs and agencies that supported these platforms, and the principal objectives of their measurements. The following subsections describe the principal goals and resources contributed by the independent programs. Appendices A and B give more experimental details of the individual platforms and sites. Figure 1 indicates the time periods over which the various platforms and sites operated.

[16] In addition to the research that is described in this special section, the ICARTT consortium also included three field studies that plan publication elsewhere. These are (1) Intercontinental Chemical Transport Experiment–North America (INTEX-NA), a NASA supported study

designed to undertake large-scale mapping of trace gases and aerosols over North America and the Atlantic Ocean (many of their results will be included in a separate INTEX-NA/ICARTT special section in *Journal of Geophysical Research*); (2) the 2004 CO_2 Boundary-layer Regional Atmospheric Study (COBRA) that examined regional-scale budgets and forest-atmosphere exchange of CO and CO_2; and (3) the U.S. DOE-operated G1 aircraft collected data from locations downwind of urban areas, and sampled point sources for trace gases and aerosols.

3.1. NEAQS-ITCT 2004 Study (NOAA)

[17] The NOAA WP-3D and DC-3 Lidar aircraft combined with the Research Vessel *Ronald H. Brown*, the surface site at Chebogue Point, and the NOAA-DOE Cooperative Agency Radar Wind Profiler network to conduct the combined New England Air Quality Study (NEAQS) and Intercontinental Transport and Chemical Transformation (ITCT) study. The WP-3D mapped trace gases, aerosols and radiative properties over the northeastern United States, and the Lidar, deployed on a chartered DC3 aircraft, mapped the regional distribution of boundary layer ozone and aerosols over New England. *Ronald H. Brown* used both in situ and remote atmospheric sensors to examine low-altitude outflow of pollution from the northeastern United States. The Chebogue Point measurements on the southern tip of Nova Scotia, approximately 500 km downwind of the New York–Boston urban corridor, provided continuous observations at a fixed site, allowing determination of the frequency and intensity of pollution events crossing the Canadian Maritime Provinces on their way to the North Atlantic. Chebogue Point included a very comprehensive measurement set for aerosol chemical and physical properties, along with a wide range of trace gas measurements and meteorological observations. Chebogue Point was also instrumented during NARE (26 July to 3 September 1993), and as such provided a point of comparison for studying temporal changes in outflow of pollution from North America. A radar wind profiler network included eleven sites that provided information on regional-scale trajectories and transport of air masses. The science plan that describes the research aims of NEAQS-ITCT can be found at http://esrl.noaa.gov/csd/2004/2004plan.pdf.

3.2. AIRMAP Network (NOAA) and CHAiOS (NSF, NOAA)

[18] AIRMAP is a program developed at the University of New Hampshire (http://www.airmap.unh.edu) to gain an understanding of regional air quality, meteorology, and climatic phenomena in New England. The AIRMAP network consists of five long-term measurement sites for documenting and studying ozone and fine particles in the region. The continuous high-resolution nature and multiyear records are strengths of the AIRMAP data set that provide a year-to-year context for the ICARTT measurements.

[19] The Chemistry of Halogens at the Isles of Shoals (CHAiOS) study, conducted at the AIRMAP site on Appledore Island, Maine, evaluated the influence of halogen radicals on the chemical evolution of pollutant outflow along the New England coast. The study focused on (1) the influences of halogen radicals on ozone production and destruction in polluted air; (2) the influence of nocturnal radical chemistry, i.e., NO_3 and N_2O_5, on halogen levels; (3) the role of halogens in the production and chemical evolution of aerosols; and (4) the potential implications of the pollutant outflow on the chemistry in the MBL over the Gulf of Maine.

3.3. PICO-NARE (NOAA, NSF)

[20] The PICO-NARE station, located on the summit caldera of Pico Mountain in the Azores (2225 m asl, 38°28.226′ north latitude, 28°24.235′ west longitude), was established to study the composition of the lower free troposphere in the central North Atlantic region, with an emphasis on the impacts of pollution outflow from the surrounding continents [*Honrath et al.*, 2004]. The station elevation allows sampling of air in the lower free troposphere [*Kleissl et al.*, 2006]. The PICO-NARE studies have as their primary objectives to (1) determine the degree to which PICO-NARE measurements are characteristic of free tropospheric composition, by analyzing the occurrence of upslope flow events on Pico mountain; (2) use observations during frequent events of boreal biomass burning emissions transport to determine the regional impact of boreal fire emissions on ozone precursors, ozone, and aerosol black carbon; (3) characterize the transport mechanisms whereby North American anthropogenic emissions are transported to the station and to assess the importance of these lower free troposphere transport events in the context of regional ozone impacts; and (4) determine the seasonal cycle of NMHC levels and HC/HC ratios in the North Atlantic lower free troposphere to quantify the impact of individual transport events on tropospheric composition.

3.4. ITOP (NERC, DLR)

[21] The 2004 Intercontinental Transport of Ozone and Precursors (ITOP, cf. http://badc.nerc.ac.uk/data/itop/) project, involving research groups from Germany, France, and the UK, made observations of chemical processing occurring in air masses transported from the United States to Europe at both high and low levels in the troposphere. The ITOP project involved the BAE146 and Falcon aircraft and the European Lidar Network. The BAE146 was based in the Azores, the approximate midpoint point between emission studies on the U.S. eastern seaboard and observations of inflowing air to Europe. A focus of the experiment was to determine the extent to which air masses remained chemically active in the days following primary emission, and the role played by relatively stable oxidative intermediates such as PAN, organic nitrates and carbonyls in extending this activity beyond the lifetime of the initially emitted species.

[22] The Falcon aircraft operated by the Deutsches Zentrum für Luft- und Raumfahrt (DLR) operated in Europe. The study objectives included interception and measurement of urban and industrial plumes transported from the northeastern United States, of forest fire plumes originating from Canada and Alaska, of European urban plumes (e.g., London, Po Valley), and of emissions from maritime shipping (e.g., in the English Channel and North Sea). The collected data set was also used for Satellite (ENVISAT) validation.

[23] Several lidar systems capable of aerosol backscatter measurements up to at least 5 km constituted the European Lidar Network, whose goal was to identify atmospheric layers not influenced by European aerosol or ozone production. Two systems also provided ozone vertical profiles.

3.5. ITCT-Lagrangian-2K4 Experiment

[24] The goal of the ITCT-Lagrangian-2K4 Experiment is to directly observe the evolution of the aerosols, oxidants and their precursors from emission over North America, trans-Atlantic transformation and transport, and impact on aerosol and oxidant levels over Europe [*Parrish and Law*, 2003]. In practice, two or three aircraft made multiple, sequential sampling flights into the same air mass during the time required for the intercontinental transport of that air mass. This plan required the close coordination of four aircraft deployed in North America (the NOAA WP-3D and the NASA DC-8), in the mid North Atlantic (the BAe-146) and in Europe (the DLR Falcon). In addition, data from the NOAA Ozone Lidar aircraft, the PICO-NARE surface site, MOZAIC measurements on commercial aircraft, the European lidar network, and European surface sites were integrated into the analyses. Each of these platforms had its own regionally focused goals, but together they provided coverage during the complete transit of a polluted air mass across the North Atlantic. Further, this activity is of central importance for ICARTT as it served to coordinate and bring together the models and measurements, and to encourage a strong instrument intercomparison effort.

3.6. ICARTT Cloud-Aerosol Study (NSF, Environment Canada)

[25] Two aircraft, the CIRPAS Twin Otter and NRC of Canada Convair 580 were involved in the Cloud-Aerosol study. The major scientific issues centered on the relationship between cloud properties and those of the aerosols upon which the clouds are forming. Therefore this experiment represents a continuing effort to obtain detailed, in situ field data that will aid in understanding the indirect climatic effect of aerosols. In addition, there was focus on understanding the atmospheric evolution of aerosols. Specific questions included the following: (1) To what extent can observed cloud drop number concentrations be predicted by theoretical aerosol-cloud activation models, given measurements of aerosol size and composition, i.e., to what extent can aerosol-cloud drop closure be achieved? What role do aerosol organic components play in determining cloud drop number concentrations? How sensitive are predicted cloud drop concentrations to the mass accommodation coefficient of water on droplets? (2) Is there evidence of liquid-phase processing of dissolved organics leading to observed organic aerosol components? (3) What processes govern the evolution of aerosols in power plant plumes as the plumes are advected from their source to the regional atmosphere? How does this evolution differ under clean versus cloudy conditions?

3.7. ICARTT Radiation-Aerosol Study (NOAA, NASA)

[26] The Jet stream-31 (J31) aircraft flew missions over the Gulf of Maine during July and August 2004. The goal was to characterize aerosol, water vapor, cloud, and ocean surface radiative properties and effects in flights that sampled polluted and clean air masses in coordination with measurements by other ICARTT platforms, including the NOAA R/V *Ronald H. Brown*, the DC-8 and DC-3 aircraft, and the Terra and Aqua satellites. Specific science objectives of the J31 included validating satellite retrievals of AOD spectra and of water vapor columns, measuring aerosol effects on radiative energy fluxes, and characterizing cloud properties using visible and near infrared reflectance in the presence of aerosols. The broader goal of the Radiation-Aerosol Study was to produce a refined, regional-scale understanding of anthropogenic aerosol and its direct radiative impact.

4. Study Coordination

[27] Because of the scope and diverse nature of the ICARTT study, considerable coordination was required. Information concerning study planning and implementation was provided to all participants via a web site (http://esrl.noaa.gov/csd/ICARTT/). The organization, planning and implementation of the study are given on the Web site. The detailed planning was tasked to six working groups: (1) aircraft and ship coordination, (2) surface networks, (3) modeling and forecasting, (4) measurement comparison, (5) data management, and (6) international coordination. These groups developed the necessary implementation to coordinate study activities. A six-member Study Coordination Team composed of individuals representing the principal programs involved in the study provided coordination among the working groups. The planning provided by these groups was presented in a series of white papers and meetings prior to the study.

[28] During the study, participants were informed on the progress of the study, given updates on operations of the various platforms and alerted to interesting finding from measurements and model predictions on the Web site under the heading of "Field Operations." Here links were provided for access to (1) the results from the measurements made at the various field sites, on the mobile platforms and realizations of satellite data; (2) model forecasts and simulations; (3) measurement intercomparison results; (4) forecast model output comparisons and forecast model comparisons with targeted field measurements; and (5) a detailed emissions map viewer that gives the location and intensity of natural and anthropogenic emission in North America.

[29] Expanded descriptions of five activities and resources that were particularly helpful in coordinating study activities follow. They are (1) the role of model simulation and forecasting, (2) the design and implementation of the ITCT-Lagrangian-2K4 experiment, (3) the emission map viewer, (4) the measurement comparison and uncertainty determination, and (5) data management protocol.

4.1. Role of Model Simulation and Forecasting

[30] A large array of model studies accompanies the observations collected during the ICARTT-2004 experiment. These models include box model analysis of in situ photochemistry, Lagrangian transport models used in the prognosis and diagnosis of intercontinental transport, and

many three-dimensional Eulerian models spanning local to global spatial scales. The forecasts provided by these models were used extensively during the study for flight planning and event interception. In addition, they suggest interesting features of events for retrospective analysis. These simulations were often made available during the study on linked web sites that were accessible to all interested participants.

[31] The modeling results included in this special journal section can be divided into four major components: those specific to AIRMAP and CHAiOS [*Chen et al.*, 2006; *Mao et al.*, 2006; M. Chen et al., Air mass classification in coastal New England and its relationship to meteorological conditions, submitted to *Journal of Geophysical Research*, 2006; R. J. Griffin et al., Contribution of gas-phase oxidation of volatile organic compounds to atmospheric carbon monoxide levels in two areas of the United States, submitted to *Journal of Geophysical Research*, 2006], Lagrangian and global Eulerian models relevant to ITOP [*Stohl et al.*, 2004; *Methven et al.*, 2006; *Real et al.*, 2006; *Cook et al.*, 2006; J.-L. E. Attie et al., Evaluation of the MOCAGE chemistry transport model during the ICARTT/ITOP experiment, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Attie et al., submitted manuscript, 2006], those associated with ITCT-Lagrangian-2K4 (see section 4.2 below), and several regional-scale Eulerian air quality forecast models (AQFMs) participating in an informal model evaluation as part of ICARTT. This last component (described further in section 6.2 below) was specifically designed to take advantage of the various surface based and aircraft platforms within ICARTT to critically assess state-of-the-art forecast models for O_3 and aerosol. ICARTT field data also play a crucial role in the boundary condition sensitivity study of Y. Tang et al. (The influence of lateral and top boundary conditions on regional air quality prediction: A multiscale study coupling regional and global chemical transport models, submitted to *Journal of Geophysical Research*, 2006) and the O_3 forecast data assimilation study of T. Chai et al. (Four dimensional data assimilation experiments with ICARTT (International Consortium for Atmospheric Research on Transport and Transformation) ozone measurements, submitted to *Journal of Geophysical Research*, 2006). As forecasts of PM2.5 aerosol, much like ozone, become routinely available to the public, the need for accurate characterization of the various processes controlling PM2.5, and evaluations of PM2.5 forecast capabilities becomes critical. G. R. Carmichael et al. (Improving regional ozone modeling through systematic evaluation of errors using the aircraft observations during ICARTT, submitted to *Journal of Geophysical Research*, 2006) utilize the various aerosol related measurements within ICARTT to evaluate PM2.5 formation and transformation.

4.2. Implementation of the ITCT-Lagrangian-2K4 Experiment

[32] The organization and realization of ITCT-Lagrangian-2K4 comprised three steps: a review of previous results, instrument comparison activities (to ensure that measurements on the disparate platforms could be accurately integrated without confounding measurement uncertainties) and flight coordination during the field deployment. The review of previous results focused on the NARE 1997 study [*Stohl et al.*, 2004], which was conducted in the same region at a similar time of year. The instrument comparison activities (discussed further in section 4.4) were focused on six wingtip-to-wingtip flights of two aircraft that together compared measurements on all four aircraft; some of the results are reported in papers in this journal section. Flight planning was based upon trajectory forecasts by models specifically developed for the purpose [*Stohl et al.*, 2004; *Methven et al.*, 2006] and discussed in daily conference calls. Several Lagrangian opportunities were identified and aircraft successfully flown to the forecast locations of the previously sampled air masses. The results are discussed in several papers in this journal section [*Methven et al.*, 2006; *Real et al.*, 2006; *Lewis et al.*, 2006; *Cook et al.*, 2006; S. R. Arnold et al., Statistical inference of OH concentrations and air mass dilution rates from successive observations of nonmethane hydrocarbons in single air masses, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Arnold et al., submitted manuscript, 2006; Attie et al., submitted manuscript, 2006; L. K. Whalley et al., unpublished manuscript, 2006].

4.3. Emission Map Viewer

[33] The analysis of the ICARTT study was facilitated by providing participants with a common emission inventory database that could be easily accessed to help identify and quantify the impact of individual point and areas sources of natural and anthropogenic emissions. A geographic information system interface, the Emission Inventory Mapviewer (http://map.ngdc.noaa.gov/website/al/emissions), which was developed by NOAA (ESRL/CSD and NGDC) in support of the ICARTT study, provided this resource. This interface allows users to easily visualize emission inventories along with various geographic data and carry out analyses of these inventories. The Emission Inventory Mapviewer was built around the EPA's 1999 National Emission Inventory (NEI99) for anthropogenic sources and the EPA's Biogenic Emissions Inventory System version 3.11 (BEIS3.11) for natural sources, over a domain covering the continental United States, southern Canada, and northern Mexico. NEI99 emissions of NO_x, CO, VOC, SO_2, NH_3, PM2.5, and PM10 from individual or groups of point sources can be viewed and downloaded. It also displays total anthropogenic emissions of these compounds on a 4-km resolution grid, and provides a convenient analysis of the partitioning between point, mobile, and area sources in any rectangular latitude-longitude region. BEIS3.11 emissions of isoprene and terpenes, the major organic components emitted by vegetation, can be visualized for standard environmental conditions. The Mapviewer can also upload and display sample aircraft flight tracks, a useful tool for planning research studies of emission sources. A complete description of the Mapviewer's data sets is given by *Frost et al.* [2006].

4.4. Measurement Comparison and Uncertainty Determination

[34] The goal of comparison exercises for the 2004 ICARTT campaign was to create a unified observational data set from measurements acquired from multiple aircraft, ground, and ship platforms. To this end, comparisons

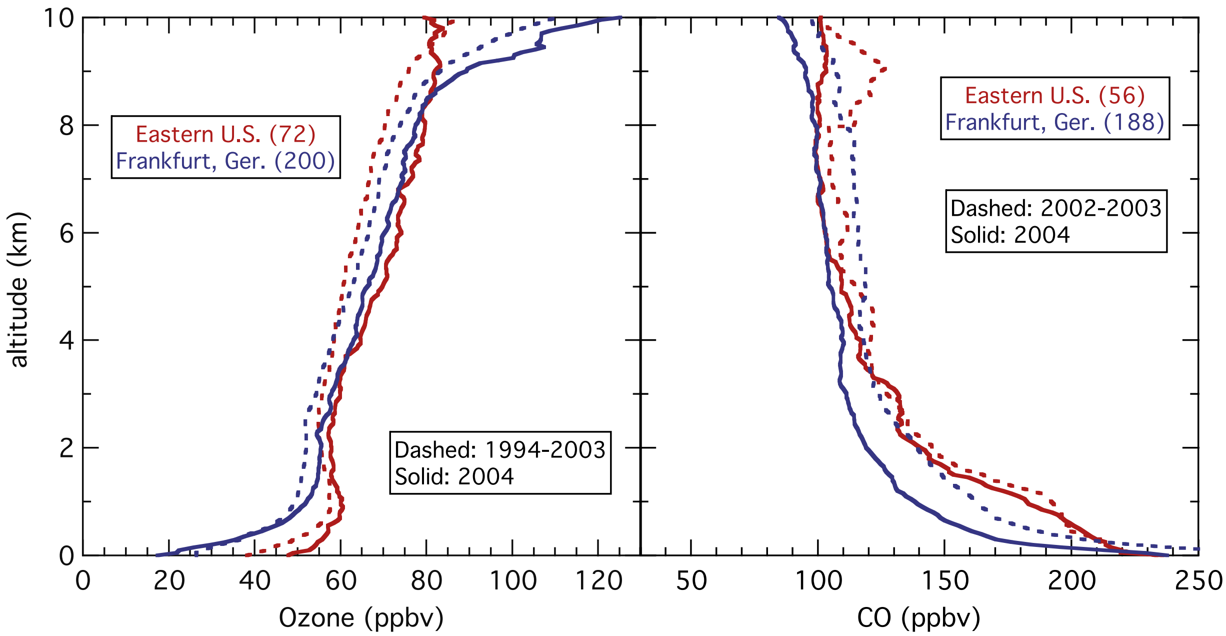

Figure 2. Average altitude profiles of ozone and CO for July–August measured by the MOZAIC program. Solid lines indicate 2004 data, and dashed lines indicate the average of all earlier years of measurements (1994–2003 for ozone and 2002–2003 for CO). Eastern U.S. represents the average from all flights into New York City, Boston, and Washington, D. C. The numbers in parentheses give the number of vertical profiles averaged in each curve for 2004.

were planned and carried out in order to help establish data comparability between the various platforms, and to verify that different analytical approaches are mutually consistent within quantifiable uncertainties. The measurements included a wide variety of in situ and remotely sensed gas-phase chemical species, aerosol chemical and physical data, radiative effects, and meteorological parameters. These data were acquired using a variety of techniques, each with specified instrumental accuracy and precision. Quantifying data uncertainty established an objective basis upon which subsequent scientific interpretations are founded.

[35] The effort required coordination between the multiple participating organizations of ICARTT, and primarily involved side-by-side measurement opportunities between combinations of aircraft, ship, and ground stations located in and between North America and Europe. In particular, comparison opportunities linked the platforms participating in the ITCT-Lagrangian-2K4 described in section 4.2. Additional comparisons of these data sets to satellite retrievals and model output are ongoing and the analyses involve the entire 2004 data set. The protocol for acquiring, evaluating, and disseminating the results of side-by-side data comparison activities for all participating platforms exclusive of satellite and model data can be found at http://esrl.noaa.gov/csd/ICARTT/.

4.5. Data Management Protocol

[36] The ICARTT study involved a large number of measurement platforms that collected a large volume of data. Each of the several laboratories involved in the study had its own procedures for handling data, so it was necessary to identify a common procedure prior to the study. This facilitated data transfer both during the study and, more importantly, after the campaign was completed. All of the principals in the study agreed upon a data transfer and archiving standard modeled after the NASA Ames format, which was chosen because it satisfied the identified data handling issues, and is easily handled by most computer-based data manipulation programs. The specifications for the Ames file exchange format can be found at http://cloud1.arc.nasa.gov/solve/archiv/archive.tutorial.html. Each group designated a data manager (listed at http://esrl.noaa.gov/csd/ICARTT/studycoordination/wgdmcontactlist.pdf) who was responsible for ensuring that all data from that group were available on an accessible server in the common format. These separate data servers (listed at http://esrl.noaa.gov/csd/ICARTT/studycoordination/wgdm.shtml) are operated and maintained by each group, and can be accessed either via the Web or ftp. Collectively, these servers constitute a distributed data repository, so no central data collection and distribution server exists.

5. Meteorological and Precursor Context of ICARTT

[37] The meteorology that prevails during a field campaign generally exerts a profound effect upon the resulting data set, and thus that data set must be interpreted within that meteorological context. Meteorology strongly affects transport patterns and stagnation conditions as well as important parameters such as radiation intensity and ambient temperature. It also affects the precursor emissions, both local (e.g., biogenic and anthropogenic hydrocarbons) and more remote (e.g., boreal forest fire emissions.)

[38] Figure 2 provides a larger spatial and temporal context for the 2004 ozone and CO distributions measured

in the ICARTT region. It presents the 2004 ozone and CO vertical profiles measured by the MOZAIC program over the eastern United States and over Germany, and compares those measurements with an average of all MOZAIC measurements from previous years. Compared to the longer-term averages, in 2004 ozone concentrations in the free troposphere were 5 to 10 ppbv higher over the eastern United States and about 5 ppbv higher over Germany. In contrast, CO was approximately 10 ppbv lower over the eastern United States and about 20 ppbv lower over Germany. During ICARTT a great deal of attention was focused on the large boreal forest fires (5.8 million hectares (A. Petzold et al., unpublished manuscript, 2006)) in Alaska and northwest Canada, which would be expected to raise the 2004 ambient CO to above normal concentrations. However, 2002 and 2003 were years of even larger-scale fires in Siberia (7.5 and 14.5 million hectares, respectively) [*Mollicone et al.*, 2006], which likely account for the higher CO concentrations throughout the midlatitude northern hemisphere in those years. It is notable that even though 2004 was characterized by few ozone extremes over the northeastern United States (see next paragraph), the average ozone profiles are not particularly low; Figure 2 shows that the 2004 ozone concentrations were higher than the preceding 10 year average, even in the continental boundary layer, at least as sampled by the MOZAIC aircraft. Interannual variability in the prevalence of key regional flow patterns perhaps could also contribute to these differences. However, *Honrath et al.* [2004] reach similar conclusions regarding the influence of the magnitude of biomass burning on the interannual variability of hemispheric CO levels.

[39] *White et al.* [2006a] have evaluated the meteorology that impacted the New England area during ICARTT. This part of the study region is particularly important because it is the "tail pipe" of North America in the sense that many of the polluted air masses leaving North America pass through this region. Thus the source for long-range transport into the North Atlantic and across to Europe may be sampled in this region. *White et al.* [2006a] contrast the July–August 2004 ICARTT period with the July–August 2002 period during which the NEAQS 2002 study was conducted in the same region. They show that these 2 years represent extremes for the 1996–2005 decade, both in meteorological conditions and in observed ozone levels. July–August 2004 accounted for the minimum number of ozone exceedences in the New England, while 2002 had the maximum. Both studies were conducted under meteorological extremes by some measures: 2002 was much warmer and drier than normal and 2004 was appreciably cooler and wetter than normal. *White et al.* [2006a] attribute the ozone extremes to these meteorological extremes. In contrast, southwesterly flow (which is most closely associated with high-pollution events) was actually more prevalent in 2004 than in 2002, and frequency of cold front passage (which is associated with disruption of pollution accumulation) was similar in the 2 years.

6. Overview of Results

[40] This section highlights some of the key findings of the ICARTT study, describes how individual results tie together, and directs interested readers to specific papers for more extensive discussions.

6.1. Air Quality: Instruments, Measurements, and Observational Based Analyses

6.1.1. Instruments

[41] The 2004 ICARRT study represented the first deployment of several significant newly developed instruments. For the first time the NOAA WP-3D aircraft carried a cavity ring-down spectroscopy (CARDS) system for simultaneous measurement of NO_3 and N_2O_5 with 1-s temporal resolution [*Dubé et al.*, 2006], a chemical ionization mass spectrometer (CIMS) instrument for the measurement of NH_3, also with 1-s resolution [*Nowak et al.*, 2006], a pulsed quantum cascade laser spectrometer for formaldehyde and formic acid [*Herndon et al.*, 2006], a Particle-into-Liquid Sampler (PILS) coupled to a Total Organic Carbon (TOC) analyzer for 3-s integrated measurements of water-soluble organic carbon (WSOC) ambient aerosol [*Sullivan et al.*, 2006], and a visible-ultraviolet spectroradiometer system for the measurement of the photolysis rate of NO_3 (H. Stark et al., Atmospheric in situ measurement of nitrate radical (NO_3) and other photolysis rates using spectro- and filter radiometry, submitted to *Journal of Geophysical Research*, 2006, hereinafter reffered to as Stark et al., submitted manuscript, 2006). The *Ronald H. Brown* Research Vessel carried a newly developed CARDS system for the measurement of aerosol extinction and a CARDS system for the measurement of NO_2 as well as NO_3 and N_2O_5 [*Osthoff et al.*, 2006b], and multisensor wind profiling system that combined a radar wind profiler, a high-resolution Doppler LIDAR, and GPS rawinsondes [*Wolfe et al.*, 2006]. The wind profiler, system provided continuous hourly wind profiles at 60- and 100-m vertical resolutions to 3–5 km height. A Thermal desorption Aerosol GC/MS-FID (TAG) instrument deployed at the Chebogue Point site reports the first ever hourly in situ measurements of speciated organic aerosol composition (B. J. Williams et al., Chemical speciation of organic aerosol during ICARTT 2004: Results from in situ measurements, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Williams et al., submitted manuscript, 2006). At the remote PICO-NARE site D. Helmig et al. (unpublished manuscript, 2006) deployed a completely automated and remotely controlled gas chromatograph for the measurement of C_2–C_6 NMHC. The system used minimal power, prepared all consumable gases and blank air at the site, and required no cryogens. O. Pikelnaya et al. (Validation of multiaxis DOAS measurements in the marine boundary layer, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Pikelnaya et al., submitted manuscript, 2006) deployed a MultiAxis Differential Optical Absorption Spectroscopy (MAX-DOAS) instrument at a surface site in the Gulf of Maine to make trace gas measurements simultaneously with long-path DOAS measurements.

6.1.2. Role of Nitrate Radicals and N_2O_5

[42] NO_3 and N_2O_5 are important atmospheric species that control nighttime chemistry. *Brown et al.* [2006a, 2006b] made the first airborne measurements of NO_3 and N_2O_5 from the NOAA WP-3D aircraft. The nocturnal concentrations of NO_3 were much larger aloft than at the

surface, and therefore far more effective at oxidizing reactive VOC, consistent with previous suggestions from models and lower-resolution determinations by remote sensing techniques. They also performed the first direct measurements of the reaction of N_2O_5 with aerosol particles. Its rate showed surprising variability that depended strongly on aerosol composition, particularly sulfate content. The correlation with aerosol composition provides evidence for a link between aerosol and ozone that is larger than previously recognized. The results have implications for the quantification of regional-scale ozone production and suggest a stronger interaction between anthropogenic sulfur and nitrogen oxide emissions than previously recognized.

[43] Simultaneous, in situ measurements were made of NO_3, N_2O_5, dimethyl sulfide (DMS), and aerosol properties from the NOAA research vessel *Ronald H. Brown* off the New England Coast during the summer of 2002 [*Stark et al.*, 2006]. Comparison between model and observed diurnal profiles of DMS and NO_3 shows that between 65 and 90% of the DMS oxidation was due to NO_3. The results have implications for the yield of sulfate aerosol from marine DMS emissions in areas affected by anthropogenic NO_x pollution. *Aldener et al.* [2006] discuss the loss of NO_3 and N_2O_5 to aerosol in the polluted marine boundary layer.

[44] The importance NO_3 and N_2O_5 during the day is usually small, but it is not always negligible. *Brown et al.* [2005] present daylight observations of both compounds from the NOAA WP-3D aircraft. The observations imply that the loss of ozone through photolysis of NO_3 to NO + O_2, oxidation of biogenic VOC, and conversion of NO_x to HNO_3 via N_2O_5 hydrolysis can be significant. *Osthoff et al.* [2006b] measured N_2O_5 from the NOAA research vessel *Ronald H. Brown* in 2004, and demonstrate that NO_3 is an important daytime oxidant for DMS, terpenes, and some anthropogenic NMHC in the polluted marine boundary layer. In foggy or hazy conditions, heterogeneous loss of N_2O_5 may be a significant NO_x sink compared to OH + NO_2.

6.1.3. Role of Halogen Radicals

[45] The CHAiOS study, conducted at Appledore Island, Maine, focused on the role of halogen radicals in tropospheric chemistry. Y. Zhou et al. (Bromoform and dibromomethane measurements in the seacoast region of New Hampshire, 2002–2004, submitted to *Journal of Geophysical Research*, 2006) report bromoform ($CHBr_3$) and dibromomethane (CH_2Br_2) measurements, and discuss their implications for the sources of these species. W. C. Keene et al. (Inorganic chlorine and bromine in coastal New England air during summer, submitted to *Journal of Geophysical Research*, 2006) found that production from sea salt was the primary source for inorganic Cl and Br species in the atmosphere even though sea-salt mass averaged 4 to 8 times lower than that typically observed over the open North Atlantic Ocean. A. A. P. Pszenny et al. (Estimates of Cl atom concentrations and hydrocarbon kinetic reactivity in surface air at Appledore Island, Maine (USA) during ICARTT/CHAiOS, submitted to *Journal of Geophysical Research*, 2006) estimated chlorine atom concentrations from variability-lifetime relationships for selected nonmethane hydrocarbons.

6.2. Air Quality: Meteorological and Modeling Studies

6.2.1. Marine Boundary Layer Characterization

[46] A shallow (≈50 m), stable boundary layer is ubiquitous over the cool waters of the Gulf of Maine in summer. This layer affects pollutant transport throughout the region by isolating overlying flow from the surface. In particular, emissions from the urban corridor of the northeastern United States can be efficiently transported long distances [*Neuman et al.*, 2006]. Transport as far as Europe in the lower troposphere has been observed. *Angevine et al.* [2006] find that the temperature profile of the lowest 1–2 km of the atmosphere over the Gulf of Maine is remarkably similar regardless of transport time over water or the time of day when the flow left the land, provided only that the flow is offshore. *Fairall et al.* [2006] find that the stable boundary layer significantly suppresses the transfer coefficients for momentum, sensible heat, and latent heat between the ocean and the atmosphere. Their estimate for the mean ozone deposition velocity corresponds to a boundary layer removal timescale of about one day. Such a short lifetime of ozone in the marine boundary layer significantly complicates the interpretation of surface ozone measurements in this marine environment.

6.2.2. Trajectory Calculations

[47] Both backward and forward air parcel trajectory calculations are important tools for investigating atmospheric transport. *White et al.* [2006b] present a trajectory calculation tool based on the radar wind profiler network observations. The continuous profiler observations allow the trajectory tool to capture changes in transport associated with mesoscale and synoptic weather events that occur between the twice-daily operational balloon soundings, thereby providing a more accurate depiction of the horizontal transport over the Gulf of Maine.

6.2.3. Model Prediction of Cloud Liquid Water Content

[48] J. Zhang et al. (Evaluation of modeled cloud properties against aircraft observations for air quality applications, submitted to *Journal of Geophysical Research*, 2006) used measured liquid water contents (LWC) in a variety of clouds to compare with values predicted from the Canadian meteorological forecast model. The model predicted the vertical distribution of LWC well, but the in-cloud LWC values were overpredicted, which will impact on the chemical processing by clouds, reinforcing the question of how best to parameterize subgrid-scale cloud processing.

6.2.4. Effect of Reductions in NO_x Emissions From Power Plants

[49] *Frost et al.* [2006] studied recent decreases in NO_x emissions from eastern U.S. power plants and the resulting effects on regional ozone. Continuous Emission Monitoring System (CEMS) measurements indicate that summertime NO_x emission rates decreased by approximately 50% between 1999 and 2003 at the subset of power plants studied. Simulations with the WRF-Chem regional chemical forecast model provide insight into the ozone changes that can be anticipated as power plant NO_x emission reductions continue to be implemented throughout the United States.

6.2.5. Ozone and PM2.5 Model Forecasts

[50] Nine AQFMs from six research centers were operational in real time during the field study covering the eastern United States and southeastern Canada. The research groups included two NOAA facilities (the NWS/NCEP CMAQ/Eta model and the ESRL/GSD WRF-Chem model), two groups from the Canadian Meteorological Service (the operational CHRONOS, and developmental AURAMS model groups), the University of Iowa (STEM-2K3 model), and the Baron Advanced Meteorological Service (MAQSIP-RT model). Studies that summarize the 2004 O_3 forecast evaluations based on the EPA AIRNOW surface O_3 monitoring network [*McKeen et al.*, 2005] and evaluations of ensemble O_3 forecast techniques [*Pagowski et al.*, 2005, 2006] have already been published for this set of models. Three papers in this section deal specifically with improved methods for forecasting surface ozone based on model ensemble techniques. Ensemble techniques have been commonly, and successfully used to improve meteorological forecasts, but they are a newer development for air quality applications. Techniques discussed in this section are bias-corrected ensemble methods [*Wilczak et al.*, 2006] Kalman filtering (L. Delle Monache et al., unpublished manuscript, 2006) and probabilistic O_3 forecasts [*Pagowski and Grell*, 2006].

[51] Compared to ozone, real-time forecasts of PM2.5 are a more recent development. *McKeen et al.* [2006] evaluate the PM2.5 forecasts from six models (and their ensembles) that were part of the ICARTT model evaluation project. The evaluation is based on comparisons with the U.S. EPA AIRNow surface PM2.5 network, composition and aerosol size distribution measurements from the NOAA WP-3 aircraft, and composition from the U.S. EPA administered STN (Speciated Trends Network) monitors.

6.3. Aerosol Formation, Composition, and Chemical Processing

6.3.1. Tropospheric Aerosol Characterization

[52] *Murphy et al.* [2006] place the aerosol composition observed in the ICARTT campaign in the context of observations from a number of airborne and ground-based campaigns through measurements of the composition of single particles by the Particle Analysis by Laser Mass Spectrometry (PALMS) instrument. L. Ziemba et al. (Aerosol acidity in rural New England: Temporal trends and source region analysis, submitted to *Journal of Geophysical Research*, 2006) describe the bulk aerosol inorganic chemical composition in northern New England, particularly in relation to aerosol acidity. Multiphase chemistry along the New England coast was investigated at Appledore Island, which receives processed continental air masses during southwesterly and westerly flow. *Fischer et al.* [2006] investigated the behavior of nitric acid/nitrate in relation to air mass transport history and local meteorology, and A. Smith et al. (Ammonia sources, transport, transformation, and deposition in coastal New England during summer, submitted to *Journal of Geophysical Research*, 2006) performed a parallel analysis of the ammonia system.

6.3.2. Nucleation and Nanoparticle Growth

[53] L. M. Russell et al. (Nanoparticle growth following photochemical a- and b-pinene oxidation at Appledore Island during ICARTT/CHAiOS 2004, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Russell et al., submitted manuscript, 2006) frequently observed nanoparticle growth events in particle size distributions measured at Appledore Island. Many of the events occurred during the morning when plentiful α- and β-pinene and ozone made production of condensable products of photochemical oxidation probable. *Ziemba et al.* [2006] present observations of frequent aerosol nucleation events in northern New England. These events were photochemically driven, most common in winter and spring, and may be associated with oxidation products of biogenic compounds, ternary homogeneous nucleation involving SO_2, and iodine chemistry from marine sources.

6.3.3. Marine Aerosol Evolution

[54] Measurements in the marine boundary layer over the Gulf of Maine from the R/V *Ronald H. Brown* were used to study the evolution of aerosols as they were transported away from the continental source regions. As distance from the source region increased, the aerosol measured in the marine boundary layer became more acidic, had a lower particulate organic matter (POM) mass fraction, and the POM became more oxidized. The POM was predominantly of secondary anthropogenic origin [*Quinn et al.*, 2006]. The relative humidity dependence of light extinction reflected the change in aerosol composition being lower for the near-source aerosol and higher for the more processed aerosol [*Quinn et al.*, 2006; Wei et al., Aerosol optical properties along the northeast coast of North America during NEAQS-ITCT 2004 and the influence of aerosol composition, submitted to *Journal of Geophysical Research*, 2006]. The aerosol light absorption to extinction ratio also changed with distance from the sources [*Sierau et al.*, 2006].

[55] J. D. Allan et al. (unpublished manuscript, 2006) and Williams et al. (submitted manuscript, 2006) characterized aerosols at Chebogue Point. The fine particulate matter was principally secondary in nature; that within plumes from the eastern United States was mainly composed of acidic sulfate and highly oxidized organics, while that from more northerly regions was mainly organic and less oxidized.

[56] Both anthropogenic and biogenic sources affected gas and particle organics at Chebogue Point. Anthropogenic and oxygenated volatile organic compounds accounted for the bulk of the gas-phase organic carbon under most conditions; however, biogenic compounds were important in terms of chemical reactivity [*Millet et al.*, 2006; R. Holzinger et al., Emission, oxidation, and secondary organic aerosol formation of volatile organic compounds as observed at Chebogue Pt, Nova Scotia, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Holzinger et al., submitted manuscript, 2006]. A suite of related oxygenated VOCs (including acetic acid, formaldehyde, acetaldehyde, formic acid and hydroxyacetone) were shown to be related to chemical species in aerosols. The compounds match the oxidation products of isoprene observed in smog chamber studies, and appear to be formed in parallel with biogenic secondary organic aerosol (Holzinger et al., submitted manuscript, 2006). Organic aerosol mass was highest during U.S. pollution events, but made up the largest

fraction of the total aerosol during biogenic oxidation events arriving from Maine and Canada [*Millet et al.*, 2006; Williams et al., submitted manuscript, 2006; J. D. Allan et al., unpublished manuscript, 2006]. In addition to anthropogenic northeastern U.S. sources, hourly measurements of particulate organic marker compounds identified several other source types, including particles formed from isoprene oxidation, particles formed from local terpene oxidation, locally produced aerosol containing large alkanes, and locally produced aerosol apparently originating from marine or dairy processing sources (Williams et al., submitted manuscript, 2006).

6.3.4. Continental Aerosol Evolution

[57] Several studies focused on the formation, growth and chemical evolution of continental aerosols. *Fountoukis et al.* [2006] measured aerosol size distributions in a power plant plume. Under both clear and cloudy sky conditions ultrafine particles grew appreciably during transport, accompanied by a decrease in the aerosol hygroscopicity. This growth and evolution may be the result of the partitioning of ambient volatile organic compounds or their oxidation products into the particle phase. *Sorooshian et al.* [2006b] provide evidence for aqueous-phase production of oxalic acid. The highest mass loadings for oxalate were measured for total aerosol and droplet residual samples in clouds influenced by power plant plumes. A chemical cloud parcel model [*Ervens et al.*, 2004] accurately predicted the relative magnitudes of the observed oxalic acid and SO_4^{2-} production. Agreement between measurements and predictions for the growth of glyoxylate, malonate, pyruvate, and glutarate provides evidence for aqueous-phase processing of dissolved organic gases contributing to aerosol organic constituents. K. L. Hayden et al. (unpublished manuscript, 2006) study changes in the partitioning of nitrate from precloud to postcloud as a function of particle size. A. Leithead et al. (unpublished manuscript, 2006) examined the airborne measurements of seven carbonyl species in cloud-water together with concurrent gas phase formaldehyde measurements, and conclude that surface adsorption and reactions, including polymerization, may contribute to the relatively high aqueous-phase levels.

6.3.5. Aerosol Organic Carbon Characterization

[58] The organic carbon (OC) aerosol contribution was a particular focus of ICARTT. *Sullivan et al.* [2006] identified two main sources of water-soluble organic carbon (WSOC) over the northeastern United States and Canada: boreal forest fire emissions from the Alaska/Yukon region and urban emissions. The boreal fire plumes contained the highest fine particle volume and WSOC concentrations of the mission. Apart from these plumes, the highest concentrations were at low altitudes in distinct plumes of enhanced particle concentrations from urban centers. Their results suggest that WSOC in fine particles is of secondary origin, produced from anthropogenic emissions rapidly converted to organic particulate matter within $\sim$1 day. *Heald et al.* [2006] examined WSOC with the GEOS-Chem global chemical transport model to test our understanding of OC aerosol in the free troposphere. Outside of the boreal fire plumes, the model accurately reproduced the average measured concentrations. This is in contrast to model performance over the NW Pacific in spring 2001 (ACE-Asia), which underestimated OC by an order of magnitude. They note that observed WSOC aerosol concentrations decrease by a factor of 2 from the boundary layer to the free troposphere, as compared to a factor of 10 decrease for sulfur oxides, indicating that most of the WSOC aerosol in the FT originates in situ.

[59] S. Gilardoni et al. (Regional variation of organic functional groups in aerosol particles on four U.S. east coast platforms during ICARTT 2004, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Gilardoni et al., submitted manuscript, 2006) collected submicron atmospheric aerosol samples on four platforms: Chebogue Point, Appledore Island, the CIRPAS Twin Otter, and the NOAA R/V *Ronald H. Brown*. Alkanes, alkene plus aromatic, organic sulfur, carbonyl and hydroxyl functional groups were measured by calibrated Fourier Transform Infrared (FTIR) spectroscopy. The functional group composition shows significant differences across the ICARTT region, with each site showing characteristic fractions of unsaturated and oxygenated carbon.

6.4. Aerosols as CCN

[60] Three cloud condensation nucleus (CCN) closure experiments were carried out using data sets collected during ICARTT in very different environments [*Fountoukis et al*, 2006; *Medina et al.*, 2006; B. Ervens et al., Prediction of CCN number concentration using measurements of aerosol size distributions and composition and light scattering enhancement due to humidity, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Ervens et al., submitted manuscript, 2006]. Each of the three experiments found excellent agreement between measured and modeled CCN concentrations, and each concluded that organic carbon does not contribute substantial amounts of solute to affect CCN activation. This supports the notion that concentrated, oxygenated organic aerosol is effectively insoluble under subsaturated conditions.

[61] The CIRPAS Twin Otter sampled highly polluted clouds within the vicinity of power plant plumes in the midwestern United States [*Fountoukis et al.*, 2006]. The uncertainty in closure between predicted and observed cloud droplet concentrations was most sensitive to updraft velocity.

[62] *Medina et al.* [2006] measured CCN, aerosol size distribution and chemical composition at the rural Thompson Farm site. The CCN closure from ”simple” Köhler theory was generally no as good during periods of changing wind direction, suggesting that introduction of aerosol mixing state would further improve closure. *Sotiropoulou et al.* [2006] used the *Medina et al.* [2006] treatment, coupled with the *Fountoukis and Nenes* [2005] activation parameterization, to evaluate the importance of CCN predictions for aerosol “indirect effect” assessments. A. Nenes and J. Medina (manuscript in preparation, 2006), using a Scanning Mobility CCN Analysis (SMCA) measurement technique, obtained high-resolution size-resolved CCN measurements at Thompson Farm during ICARTT. SMCA provides insight into the chemical composition of the aerosol, as well as detailed information on the CCN mixing state and size-resolved droplet growth kinetics.

[63] Ervens et al. (submitted manuscript, 2006) predicted the number concentration of CCN from measurements of

aerosol size distribution, composition, and hygroscopic growth made at Chebogue Point, Nova Scotia (a marine rural site receiving well aged air masses). They show that CCN can be predicted quite reliably using measured size distributions, a simple soluble/insoluble aerosol model, and either the diameter growth factor $g(RH)$ or the light scattering growth factor $f(RH)$.

[64] *Garrett et al.* [2006] provide a measurement technique for assessing the extent to which concentrations of CCN and HNO_3 are scavenged by precipitation, distinct from the separate sinks of dilution, dry deposition, and chemical transformation. The technique does not require detailed knowledge of the aerosols, clouds and precipitation involved, only measurements of HNO_3 and CO in clear air. This technique may provide a method for evaluating parameterizations of chemical and aerosol sinks parameterized in transport models.

6.5. Aerosol Radiative Effects

[65] The Jet stream 31 aircraft flew over the Gulf of Maine to characterize aerosol, water vapor, cloud, and ocean surface radiative properties and effects in flights that sampled polluted and clean air masses in coordination with measurements by other ICARTT platforms, including several satellites. *Redemann et al.* [2006] report measurements of aerosol effects on radiative energy fluxes. They found a high variability in the aerosol forcing efficiencies for the visible wavelength range, and derive 24-hour-average values for the forcing efficiency.

[66] *Avey et al.* [2006] characterize the "indirect effect" of pollution aerosol on clouds and climate using combined satellite retrievals of clouds and aerosols. Aircraft data indicate that measured CO perturbations (used as a pollution tracer) correspond to smaller measured values of cloud droplet effective radii, r_e, and higher droplet number concentrations. Satellite data show that mean values of retrieved r_e are smaller under modeled polluted conditions.

6.6. Long-Range Transport

6.6.1. North American Outflow

[67] The surface site operated at Chebogue Point sampled surface outflow from the eastern seaboard of North America. Three-dimensional chemical transport model results show that Chebogue Point is well situated to sample surface layer pollution outflow. However, 70% of the export takes place above 3 km, so that aircraft and satellite observations are also needed to fully characterize North American outflow. The overall distributions of ozone and CO in air arriving at Chebogue Point were very similar in 1993 and 2004 [*Millet et al.*, 2006]. Measured particulate matter within plumes from the eastern United States was principally secondary in nature, mainly composed of acidic sulfate and highly oxidized organics (Williams et al., submitted manuscript, 2006; J. D. Allan et al., unpublished manuscript, 2006).

[68] The NOAA WP-3 aircraft extensively studied plumes of North American emissions over the western north Atlantic. *Neuman et al.* [2006] characterize urban emissions, and plume transport and transformation processes in aged plumes located up to 1000 km downwind from the east coast of North America. Emission outflow was observed primarily below 1.5 km altitude in well-defined layers that were decoupled from the marine boundary layer. In aged plumes located over the North Atlantic Ocean, the nitric acid (HNO_3) mixing ratios were large (up to 50 ppbv) and HNO_3 accounted for the majority of reactive nitrogen. Plume CO and reactive nitrogen enhancement ratios were nearly equivalent in fresh and aged plumes, which indicated efficient transport of HNO_3. Without substantial HNO_3 loss, the ratio of HNO_3 to NO_x was between 13 and 42 in most highly aged plumes and sometimes exceeded calculated photochemical steady state values, which indicate the contribution of nighttime reactions in the conversion of NO_x to HNO_3. Photolysis and OH oxidation of over 10 ppbv HNO_3 that was in the troposphere for days resulted in reformation of hundreds of pptv of NO_x, which is sufficient to maintain photochemical ozone production. The efficient transport of HNO_3 carried both HNO_3 and NO_x far from their sources, extended their atmospheric lifetimes, and increased their photochemical influence.

[69] *Parrish et al.* [2006] describe a model for investigating the combined influences of photochemical processing and air mass mixing on the evolution of nonmethane hydrocarbon (NMHC) ratios. The model-measurement comparisons indicate that the interaction of mixing and photochemical processing prevent a simple interpretation of "photochemical age," but that the average age of any particular NMHC can be well defined, and can be approximated by a properly chosen and interpreted NMHC ratio. The relationships of NMHC concentration ratios not only yield useful measures of photochemical processing in the troposphere, but also provide useful tests of the treatment of mixing and chemical processing in chemical transport models.

6.6.2. Lagrangian Balloon Systems

[70] During the ICARTT campaign, altitude-controlled balloons tracked urban pollution plumes. Nine balloons flew a total of 670 flight hours, measuring the quasi-Lagrangian evolution of the winds, temperature, and ozone downwind of major pollution source regions and helping mission scientists to find the emission plumes in real time. Two types of balloons were flown: NOAA's SMART balloons [*Mao et al.*, 2006], released from the eastern tip of Long Island in New York, with one flight reaching Europe. Smaller Controlled Meteorological (CMET) balloons [*Riddle et al.*, 2006] were launched from multiple locations in order to target specific plumes. Flights ranged from 12 to 120 hours in duration. Mean trajectory errors were found to be approximately 25% of the flight distance for ECMWF-based trajectories.

6.6.3. Mid-Atlantic Environment

[71] The PICO-NARE site has provided unprecedented measurements in the free troposphere in the most remote part of the central North Atlantic Ocean. The year-round data elucidate seasonal cycles of tropospheric chemistry in this region with good statistics from relatively long-term measurements. Emissions from North American boreal fires frequently reached the PICO-NARE station during summer 2004, significantly increasing levels of nonmethane hydrocarbons (NMHC) (D. Helmig et al., unpublished manuscript, 2006), nitrogen oxides, carbon monoxide and black carbon, and increasing ozone as well in most cases [*Val Martín et al.*, 2006]. The magnitude of the observed levels

and the distance of the Azores from the fires implies large-scale impacts of boreal fires on lower-tropospheric composition, and is consistent with multiyear analyses of correlations between upwind boreal fires and increased ozone at the station [*Lapina et al.*, 2006]. D. Helmig et al. (unpublished manuscript, 2006) utilize 1 year of continuous measurements of NMHC at the PICO-NARE station to investigate seasonal oxidation chemistry. Interpretations of NMHC ratios as a relative measure of photochemical processing indicate that in spring enhanced ozone levels were observed in air that had relatively "fresh" photochemical signatures and ozone at lower levels was observed in more processed air. This relationship indicates that the lower troposphere over the central North Atlantic is a region of net ozone destruction in spring.

6.6.4. Low-Level Anthropogenic Pollution Outflow

[72] *Owen et al.* [2006] analyze low-level transport events that brought North American anthropogenic emissions to the PICO-NARE station. Low-level transport during summer 2003 resulted in frequent CO enhancements at the station. Although exported and transported at low altitudes, these events were observed at 2.2 km, well above the marine boundary layer, and were characterized by significant enhancements in ozone. These ozone enhancements may reflect the efficient transport of nitric acid in plumes above the marine boundary layer [*Neuman et al.*, 2006]. *Owen et al.* [2006] suggest that transport in the lower free troposphere above the marine boundary layer, may provide an effective mechanism for long-range impacts of anthropogenic emissions on lower-tropospheric ozone in distant downwind regions.

6.6.5. PICO-NARE Station Future

[73] Research described in this section has contributed to the continuation of active measurements at the PICO-NARE site, which was originally installed as a temporary research station but is now the focus of development of a permanent Portuguese observatory. *Kleissl et al.* [2006] determined that measurements there are usually characteristic of the free troposphere, even during summer (when buoyant upslope flow affects the station much less frequently than it does many other mountaintop observatories). This is the result of the latitude, small size, and topography of Pico Mountain. The station is valuable for the observation of highly aged but detectable plumes of anthropogenic [*Owen et al.*, 2006] and boreal forest fire [*Val Martín et al.*, 2006] plumes, and provides a platform for year-round observations characteristic of regional background levels, as demonstrated for NMHCs by D. Helmig et al. (unpublished manuscript, 2006).

6.6.6. Impact in Inflow Regions

[74] The final destination of a significant fraction of the emissions that have been transported over long distances is arrival over downwind continental regions, where they can be entrained into the continental boundary layer and affect the air quality of those regions. The ICARTT program in general and the ITCT-Lagrangian-2K4 study in particular were designed to evaluate the impact of North American emissions on Europe; however, the impact of intense Alaskan and Canadian boreal forest fires were also noted at distant locations in North America.

[75] A. Petzold et al. (unpublished manuscript, 2006) use the data collected during ICARTT study and combine it with data from two ground sites in Central Europe to investigate the influence of the boreal fire smoke layers on the aerosol properties in the free troposphere and the continental boundary layer of Central Europe. F. Ravetta et al. (Impact of long-range transport on tropospheric ozone variability in western Mediterranean region during ITOP-2004, submitted to *Journal of Geophysical Research*, 2006) used lidar measurements in Europe to link ozone rich layers within the free troposphere to long-range transport of pollutants. These layers had their origin in North America where they were uplifted either by forest fires or by warm conveyor belts in the vicinity of frontal regions. The polluted layers remained coherent during transport over the Atlantic Ocean. T. J. Duck et al. (Transport of forest fire emissions from Alaska and the Yukon Territory to Nova Scotia during summer 2004, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Duck et al., submitted manuscript, 2006) report aerosol lidar observations at Chebogue Point, Nova Scotia, which indicate transport of a boreal forest fire plume from Alaska to the site, where the plume was brought from the free troposphere to the surface by synoptic-scale meteorology.

[76] The NOAA WP-3 aircraft intercepted aged boreal forest fire plumes from Alaska and northwest Canada over the New England area [*de Gouw et al.*, 2006]. The removal of aromatic VOCs was slow, implying that the average OH concentrations were low during the transport. Low humidity and high concentrations of carbon monoxide and other pollutants account for the low OH concentrations in the plumes. In contrast with previous work, no strong secondary production of acetone, methanol and acetic acid were inferred from the measurements. A clear case of removal of submicron particle volume and acetic acid due to precipitation scavenging was observed. *Warneke et al.* [2006] conducted a source apportionment study of CO downwind of the Boston–New York City urban complex, and find that as much as 30% of the measured CO enhancement is attributed to the forest fires in Alaska and Canada transported into the region.

6.7. ITCT Lagrangian 2K4 Related Studies

[77] In the ITCT-Lagrangian-2K4 study a combination of trajectory analyses and independent chemical signatures was used to establish the occurrence of events where chemical processing could be studied in a Lagrangian framework on intercontinental scales. *Methven et al.* [2006] provide evidence that this type of experiment has for the first time been successfully achieved in the free troposphere.

[78] Analysis of identified Lagrangian events on the North America to Europe intercontinental scale allowed investigation of the chemical environment of the mid Atlantic. For the most part a small tendency for net ozone production with a concurrent loss of CO was identified [*Methven et al.*, 2006]. A major feature of the ICARTT study period was the strength and importance of low-level (below 700 hPa) transport of continental emissions at altitudes just above the marine boundary layer. (This transport is in addition to the expected transport pathways in the mid and upper troposphere.) This feature is in particular contrast to previous ACSOE aircraft studies made in 1997,

also based in the Azores. A consequence of the low-level transport was the elevation in NO available in the lower troposphere in the mid Atlantic (both from direct transport and *via* decomposition of sequestered forms), with notable impacts on calculated ozone production efficiency in this region [*Lewis et al.*, 2006]. The evolution of nonmethane hydrocarbons (NMHC) between the interceptions in the Lagrangian events was exploited by Arnold et al. (submitted manuscript, 2006) to estimate the mean OH concentrations and dilution rates acting over the time intervals between observations. These are the first estimates of time mean OH concentrations following individual air masses over several days, which are well constrained by observations up and downwind.

[79] The interception of biomass burning plumes several thousand kilometers downwind of aircraft observations near North America indicate that mixing was often very limited between the stretching filaments and the background. Tracers such as CO reached concentrations as high as 600 ppbv in biomass burning plumes intercepted in the mid Atlantic, similar values to those seen much closer to source, and within these air masses there remained a significant distribution of reactive chemicals, notably the elevation of the unsaturated hydrocarbon ethene [*Lewis et al.*, 2006]. However, when the filaments reached frontal boundaries, mixing produced more pronounced effects [*Real et al.*, 2006].

[80] *Real et al.* [2006] analyze in detail one case of long-range transport of a biomass burning plume from Alaska to Europe. This plume was sampled several times in the free troposphere over North America, the North Atlantic and Europe by three different aircraft. The measurements showed enhanced values of CO, VOCs and NO_y, primarily in the form of peroxyacetylnitrate (PAN), and the measured ozone increased by 17 ppbv over the 5 days of transport from North America to Europe. A photochemical trajectory model, initialized with upwind data, indicated that the large ozone increases were primarily due to PAN decomposition during descent of the plume toward Europe. The predicted ozone changes were very dependent on the temperature during transport, and on the water vapor levels in the lower troposphere, which lead to ozone destruction. Inclusion of mixing of the plume with adjacent air masses was found to be important for the model simulations to agree well with observed changes in CO and ozone. The simulated evolution of the O_3/CO correlations in the plume agreed well with observations, where the slopes changed from negative to positive over the five days of transport. The possible impact of this plume on ozone levels in the European boundary layer is also examined by extending the model for a further five days, and comparing with data collected at surface sites.

7. Conclusions

[81] The ICARTT measurements constitute a remarkably rich data set for investigating regional air quality, the transport, chemical transformation and removal of aerosols, O_3, and their precursors during intercontinental transport, and the radiation balance of the troposphere. The results presented in this special section of *Journal of Geophysical Research* represent only the initial analysis; the data set is available to the atmospheric chemistry community for further analysis in the coming years.

Appendix A: Mobile Platform Instrument Payloads and Deployment Details

[82] The NOAA WP-3D aircraft was instrumented to study aerosol composition and gas-phase chemical transformations. The aircraft operated from the PBL up to 6.4 km and had sufficient range to reach from the central-northeastern United States to the maritime Canadian Provinces, and well out into the North Atlantic while stationed at the Pease Tradeport in New Hampshire. Tables A1a and A1b summarize the characteristics of the WP-3D instrumentation, and Table A2 and Figure A1 summarize the ICARTT flights.

[83] The NOAA airborne ozone/aerosol differential absorption lidar (DIAL) [*Alvarez et al.*, 1998] was deployed on a chartered DC-3 aircraft, also stationed at the Pease Tradeport. The nadir-looking lidar measured ozone profiles in the boundary layer with high spatial resolution (90 m vertical, 600 m horizontal) with a precision that varied between 5 and 15 ppbv, depending on the total atmospheric extinction. The lidar also provided aerosol backscatter profiles with a vertical resolution of 15 m. In addition, an analyzer measured ozone at flight levels, an infrared radiometer observed surface skin temperature variations, and there were dropsonde capabilities. The DC-3 flew a total of 98 flight hours during ICARTT, in flights ranging between about 5 and 8 hours duration. The aircraft generally flew at 3 km ASL where lidar observations were obtained from 2.2 km ASL to just above the surface. Figure A2 illustrates the DC-3 flight tracks.

[84] The NOAA Research Vessel *Ronald H. Brown* conducted two 19 day cruises out of Portsmouth, New Hampshire from 5 to 23 July 2004 and 26 July to 13 August 2004. The ship was instrumented to measure an extensive set of in situ gas and aerosol parameters as well as many remotely sensed parameters (Table A3). Radiosondes (2–8 times per day) and ozonesondes (daily) also were launched from the ship. The cruise tracks in the Gulf of Maine are shown in Figure A3.

[85] ITOP provided the first science mission for the new FAAM BAE146 research aircraft, instrumented primarily for gas phase measurements, but with a limited capacity for concurrent aerosol observations. The aircraft operated within the altitude range from 50ft over the sea surface to 9 km, and spatially between 20–40°W and 33–47°N. Operations were based in Horta Airport, on Faial Island one of the Azores archipelago. Tables A4a and A4b summarize the characteristics of the BAE146 instrumentation, and Table A5 and Figure A4 summarize the ITOP flights

[86] The DLR Falcon performed the ITOP measurement flights in Europe. The missions were performed from 2 July to 3 August 2004 from the DLR airport in Oberpfaffenhofen near Munich and the airport in Creil near Paris. The aircraft has a maximum flight altitude of 41000 feet when fully instrumented including wing pods. The minimum flight altitude is 100 and 300 m over the ocean and over land, respectively. Maximum range and endurance is 3000 km and 4 hours. The measurement speed varies between 100 and 180 m s^{-1} depending on flight altitude.

Table A1a. NOAA WP-3D Aircraft Instrumentation for Gas-Phase Measurements

Species/Parameter	Reference	Technique	Averaging Time	Accuracy	Precision	Detection Limit
NO	*Ryerson et al.* [1999]	NO/O_3 chemiluminescence	1 s	5%	10 pptv	20 pptv
NO_2	*Ryerson et al.* [1999]	photolysis-chemiluminescence	1 s	8%	25 pptv	100 pptv
NO_y	*Ryerson et al.* [1999]	Au converter-chemiluminescence	1 s	10%	20 pptv	50 pptv
O_3	*Ryerson et al.* [1998]	NO/O_3 chemiluminescence	1 s	3%	0.1 ppbv	0.2 ppbv
CO	*Holloway et al.* [2000]	VUV resonance fluorescence	1 s	2.5%	0.5 ppbv	1 ppbv
H_2O		Lyman alpha absorption	1 s	. . .	. . .	. . .
H_2O		thermoelectric hygrometer	3 s	±0.2°–1.0°C	±0.2°–1.0°C	−75° to +50°C
NMHCs (C_2-C_{10})	*Schauffler et al.* [1999]	grab sample/GC	8–30 s[a]	5–10%	1–3%	3 pptv
Halocarbons (C_1-C_2)	*Schauffler et al.* [1999]	grab sample/GC	8–30 s[a]	2–20%	1–10%	0.02–50 pptv
Alkylnitrates (C_1-C_5)	*Schauffler et al.* [1999]	grab sample/GC	8–30 s[a]	10–20%	1–10%	0.02 pptv
VOCs	*de Gouw et al.* [2003]	proton transfer reaction mass spectrometer (PTRMS)	1 s every 15 s	10–20%	5–30%	50–250 pptv
Formaldehyde	*Jimenez et al.* [2005]	tunable infrared diode laser absorption spectroscopy (TIDLAS)	1 s	7%	300 pptv	140 pptv
Formic acid	*Jimenez et al.* [2005]	TIDLAS	1 s	33%	400 pptv	180 pptv
PAN, PPN, PiBN, APAN MPAN	*Slusher et al.* [2004]	chemical ionization mass spectrometer (CIMS)	2 s, 2 s	15%, 30%	2%, 2%	1 pptv, 5 pptv
NO_3, N_2O_5	*Dubé et al.* [2006]	cavity ring-down spectroscopy (CARDS)	1 s	25%	2%	1 pptv
HNO_3, NH_3	*Neuman et al.* [2002]	CIMS	1 s	15%	25 pptv	50 pptv
Hydroxyl radical	*Eisele and Tanner* [1993]	CIMS	30 s	35%	1×10^6 cm^{-3}	5×10^5 cm^{-3}
SO_2	*Ryerson et al.* [1998]	pulsed UV fluorescence	3 s	10%	0.35 ppbv	1 ppbv
H_2SO_4	*Eisele and Tanner* [1993]	CIMS	1.1 s	35%	1×10^6 cm^{-3}	1×10^6 cm^{-3}
SO_2, O_3, H_2O column		miniature differential absorption spectroscopy (MIDAS)				

[a]Dependent upon altitude.

Tables A6a and A6b compile the instrumentation used for ITOP. The instruments were provided and operated by DLR-Institute for Atmospheric Physics in Oberpfaffenhofen, the Max-Planck-Institutes for Chemistry and Nuclear Physics in Mainz and Heidelberg, respectively, and the Institute for Atmospheric Environmental Research (IFU) of the Research Center in Karlsruhe. Table A7 gives an overview of all Falcon measurement flights during ITOP including flight objectives. Missions were conducted on 11 different days, some of them including fuel stops (in Cranfield, UK, San Sebastian, Spain and Shannon, Ireland).

[87] During 2–21 August 2004, the CIRPAS Twin Otter aircraft was based at Hopkins International Airport in Cleveland, Ohio. The payload consisted of a wide array of instrumentation for aerosol cloud physical and chemical characterization, employing both online and off-line techniques (Table A8). The general focus of the mission was on characterizing aerosol and cloud droplets, from within the boundary layer up to the free troposphere. A variety of air mass types was sampled during this study, including plumes from coal-fired power plants (Conesville and Detroit Monroe plants) both in clear sky and under cloudy conditions, cloud systems over Ohio and Lake Erie, urban outflow from Detroit and Cleveland, and clear air masses on various transit legs. Table A9 lists the research flights during ICARTT, and Figure A5 shows the individual flight

Table A1b. NOAA WP-3D Aircraft Instrumentation for Aerosol and Ancillary Data Measurements

Species/Parameter	Reference	Technique	Averaging Time	Detection Limit
Aerosol single particle composition	*Thomson et al.* [2000]	particle analysis by laser mass spectrometry (PALMS)	single particle	<1 cm^{-3}
Aerosol bulk ionic composition	*Weber et al.* [2001] and *Orsini et al.* [2003]	particle into liquid sampling (PILS)–ion chromatography (IC)	3 m	<0.02 μg/m^3
Aerosol water soluble organic composition	*Sullivan et al.* [2006]	particle into liquid sampling (PILS)–total organic carbon (TOC)	3 m	0.3 μg/m^3
Aerosol nonrefractory, size-resolved composition	*Bahreini et al.* [2003]	aerosol mass spectrometer (AMS)	10 m	SO_4^{-2}, 0.1 μg/m^3; NO_3^-, 0.1 μg/m^3; NH_4^+, 0.4 μg/m^3; organics, 0.6 μg/m^3
Small aerosol size distribution	*Brock et al.* [2000]	nucleation mode aerosol size spectrometer (NMASS)	1 s	0.005–0.06 μm
Large aerosol size distribution	*Brock et al.* [2003] and *Wilson et al.* [2004]	light scattering (white light and laser) with low turbulence inlet	1 s	0.12–8.0 μm
Photolytic flux	Stark et al. (submitted manuscript, 2006)	280–690 nm spectrally resolved radiometer, zenith and nadir	1 s	2×10^{11} photons cm^{-2} s^{-1} at 500 nm
Broadband radiation		pyrgeometer	1 s	3.5–50 μm
Broadband radiation		pyranometer	1 s	0.28–2.8 μm

Table A2. NOAA WP-3D Flights

Flight	Flight Description	Date in 2004	Takeoff-Landing, UT
1	transit Tampa, Florida, to Pease Tradeport, New Hampshire	5 Jul	1610–2211
2	survey Boston urban plume, Alaskan biomass burning plumes	9 Jul	1529–2301
3	Boston urban plume at night	11 Jul	2255–0351
4	North American plume at 60°W and New York City urban plume	15 Jul	1310–2113
5	New York City urban plume: near source	20 Jul	1411–2213
6	New York City urban plume: over Gulf of Maine	21 Jul	1402–2029
7	New York City urban plume: Nova Scotia; DC-8 intercomparison	22 Jul	1348–2134
8	point source and urban plume evolution in the northeast United States	25 Jul	1415–2207
9	characterize pollution accumulation ahead of cold front	27 Jul	1503–2227
10	WCB outflow of accumulated pollution; biomass burning plumes	28 Jul	1354–2033
11	New York City urban plume at night; DC-8 intercomparison	31 Jul	2124–0516
12	New York City urban plume at night	3 Aug	0153–0823
13	Ohio River Valley power plant plumes	6 Aug	1400–2227
14	New York City, Boston urban plumes at night; DC-8 intercomparison	7 Aug	2008–0436
15	Ohio River Valley power plants, New York City urban plumes at night	9 Aug	2257–0729
16	New York City urban plume: night into day	11 Aug	0300–1051
17	cloud investigation	14 Aug	1355–2210
18	transit Pease Tradeport, New Hampshire, to Tampa, Florida, via Atlanta, Georgia	15 Aug	1434–2129

tracks. Six of the 12 flights were also flown in coordination with the MSC Convair aircraft, which enabled complementary aerosol and gas-phase measurements.

[88] The MSC Convair 580 also was based at Hopkins International Airport in Cleveland, Ohio for ICARTT from 21 July to 18 August 2004. The Convair carried instrumentation to measure or collect trace gases (O_3, CO, SO_2, NO, NO_2, HCHO, H_2O_2, HNO_3 and some VOCs), aerosol particles and cloud droplets. Both the physical size distributions and the chemistry of the aerosol particles were measured using a DMA, an APS, a PCASP, a FSSP300, a PILS and an AMS. An Alqhuist three-wavelength integrating nephelometer and a PSAP were used to measure the scattering and absorption properties of the particles. Cloud liquid water content was measured with a PMS King probe and a Nezorov probe. Cloud microphysics were measured with two PMS FSSP 100 probes, a PMS 2D Grey scale and a PMS 2DP. Light scattering by cloud droplets was measured with a Gerber CIN probe. The chemistry of the cloud droplets was measured in two ways: sampling the residuals from a CVI into the AMS, and collecting bulk samples of the cloudwater using slotted rod collectors. A total of 23 flights were conducted with the Convair. After 1 August, six flights were made in unison with the CIRPAS Twin Otter. Table A10 lists the project flights during ICARTT, and Figure A6 shows a compilation of the individual flight tracks.

[89] During the ICARTT campaign, altitude-controlled balloons were used to track urban pollution plumes. Nine balloons flew a total of 670 flight hours, measuring the evolution of the winds, temperature, and ozone downwind of major pollution source regions and helping to track

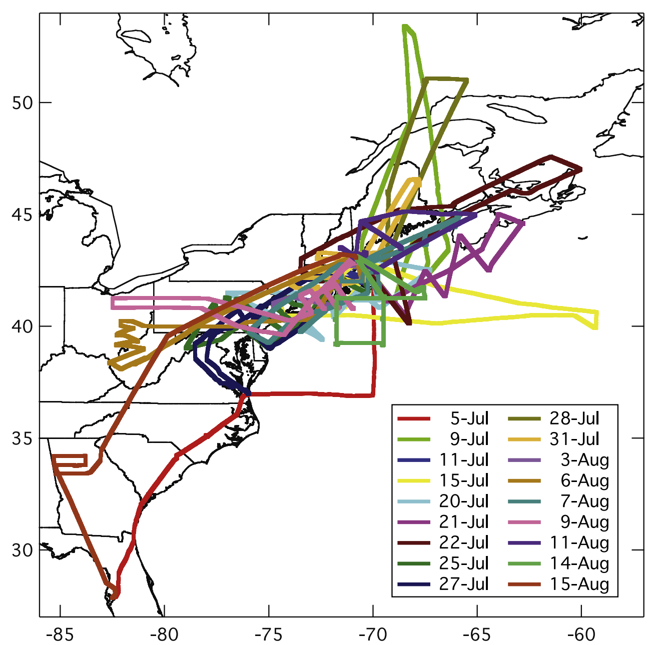

Figure A1. Flight tracks of NOAA WP-3D aircraft during ICARTT.

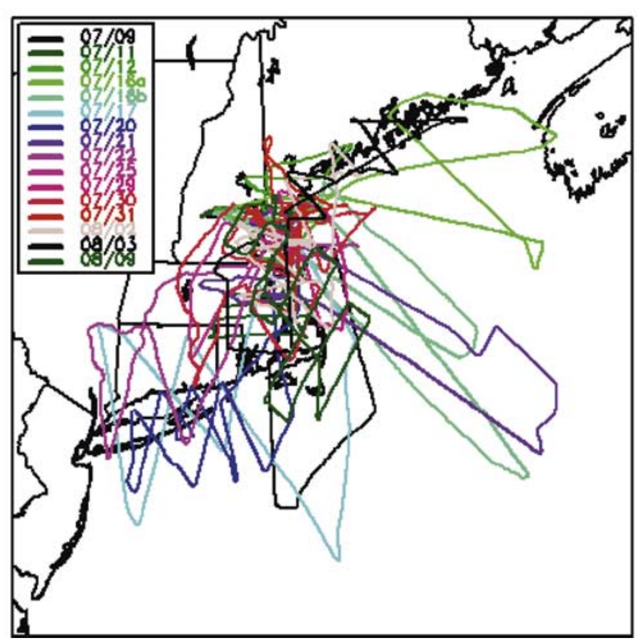

Figure A2. Flight tracks of NOAA DC-3 lidar aircraft during ICARTT.

Table A3. NOAA Research Vessel *Ronald H. Brown* Instrumentation

Species/Parameter	Reference	Technique	Averaging Time	Detection Limit	Uncertainty
JNO_2 photolysis rates	*Shetter et al.* [2003]	spectral radiometer	1 min	5e-7 Hz	±22%
JNO_3 photolysis rates	Stark et al. (submitted manuscript, 2006)	spectral radiometer	1 min	3e-7 Hz	±30%
$JO_3(^1D)$ photolysis rates	*Bohn et al.* [2004]	spectral radiometer	1 min	4e-8 Hz	±30%
Ozone	*Bates et al.* [2005]	UV absorbance	1 min	1.0 ppb	±1.0 ppb or 2%
Ozone	*E. J. Williams et al.* [2006]	NO chemiluminescence	1 min	0.1 ppbv	±(2% + 1.0 ppbv)
NO_2	*Sinreich et al.* [2005]	passive DOAS	5 min	0.1 ppb	70 ppt
CH_2O	*Sinreich et al.* [2005]	passive DOAS	5 min	0.3 ppb	0.2 ppb
BrO	*Sinreich et al.* [2005]	passive DOAS	5 min	1 ppt	0.7 ppt
Ozone vertical profiles	*Thompson et al.* [2000a]	ozonesondes	1 s = 5 m	2 ppbv	3–5%
Ozone vertical profiles	*Zhao et al.* [1993]	O_3 lidar (OPAL)	10 min	5 ppb	<10 ppb
Carbon monoxide	*Gerbig et al.* [1999]	UV fluorescence	1 min	1.0 ppb	±3.0%
Carbon dioxide	LiCor spec	nondispersive IR	1 min	0.07 ppm	±2.5%
Water vapor	LiCor spec	nondispersive IR	1 min	1 ppm	±1%
Sulfur dioxide	*Bates et al.* [2005]	pulsed fluorescence	1 min	100 ppt	<5%
Nitric oxide	*Osthoff et al.* [2006a]	chemiluminescence	1 min	18 ppt	±(4% + 7 pptv)
Nitrogen dioxide	*Osthoff et al.* [2006a]	photolysis cell	1 min	27 ppt	±(6.5% + 93 pptv) at $NO_2/NO = 3$
Total nitrogen oxides	*Williams et al.* [1998]	Au tube reduction	1 min	0.04 ppbv	±(10% + 0.08 ppbv)
PANs	M. Marchewka et al. (unpublished manuscript, 2006)	GC/ECD	1 min	PAN/PPN (5 pptv); PiBN/MPAN (10 pptv)	PAN/PPN ± (5 pptv + 15%); PiBN/MPAN ± (10 pptv + 20%)
Alkyl nitrates	*Goldan et al.* [2004]	GC/MS	5 min	≤1 ppt	±20%
NO_3/N_2O_5	*Dubé et al.* [2006]	cavity ring-down spectrometry	1 s	1 pptv	1 pptv, ±30%
NO_2	*Osthoff et al.* [2006a]	cavity ring-down spectrometry	1 s	160 pptv	160 pptv, ±8%
Nitric acid/NH_3	*Dibb et al.* [2004]	automated mist chamber/IC	5 min	5 pptv	15%
Radon	*Whittlestone and Zahorowski* [1998]	radon gas decay	13 min		
VOC speciation	*Goldan et al.* [2004]	GC/MS	5 min	≤1 ppt	±20%
Seawater and atmospheric pCO_2	*Sabine et al.* [2000]	nondispersive IR	30 min		±0.2 ppm
Seawater DMS	*Bates et al.* [2000]	S chemiluminesence	30 min	0.2 nM	±8%
Continuous speciation of VOCs	*Warneke et al.* [2005]	PTR-MS/CIMS	2 min	50–500 pptv	20%
Aerosol ionic composition	*Quinn et al.* [2006]	PILS-IC	5 min		
Aerosol WSOC	*Quinn et al.* [2006]	PILS-TOC	1 hour		
Aerosol size and composition	*Quinn et al.* [2006]	aerosol mass spectrometer	5 min	0.1 $\mu g\ m^{-3}$	±20%
Aerosol OC	*Quinn et al.* [2006]	online thermal/optical	1 hour	0.1 $\mu g\ m^{-3}$	
Aerosol organic functional groups	Gilardoni et al. (submitted manuscript, 2006)	FTIR spectroscopy of <1 μm particles on Teflon filters	4–12 hours	1 μg	±15%
Aerosol composition, 2 stage (sub/super micron) and 7 stage at 60% RH	*Quinn andBates* [2005]	impactors (IC, XRF and thermal optical OC/EC, total gravimetric weight)	4–12 hours		±6–31%
Total and submicron aerosol scattering and backscattering (450, 550, 700 nm) at 60% RH	*Quinn andBates* [2005]	TSI 3563 nephelometers (2)	1 min		±14%
Total and submicron aerosol absorption (450, 550, 700 nm) dry	*Sierau et al.* [2006]	Radiance Research PSAPs (2)	1 min		±22%
Total and submicron aerosol extinction	T. Baynard et al. (Design and application of a pulsed cavity ring-down aerosol extinction spectrometer for field measurements, submitted to *Aerosol Science and Technology*, 2006)	cavity ring-down spectrometry	1 min	0.01 Mm^{-1}	±1%

Table A3. (continued)

Species/Parameter	Reference	Technique	Averaging Time	Detection Limit	Uncertainty
Aerosol number	*Bates et al.* [2001]	CNC (TSI 3010, 3025)	1 s		±10%
Aerosol size distribution	*Bates et al.* [2005]	DMA and APS	5 min		±10%
Total and submicron aerosol light scattering hygroscopic growth	*Carrico et al.* [2003]	twin TSI 3563 nephelometers; RR M903 nephelometer	20 s (over each 1% RH)	σ_{spTSI}, 1.85 and 2.78; σ_{bsp}, 1.24 and 2.96; σ_{spRR}, 1.06	σ_{spTSI}, $-14 \sim 17$; σ_{bsp}, $-17 \sim 19$
Aerosol optical depth	*Quinn and Bates* [2005]	Microtops	intermittent		±0.015 AOD
Aerosol backscatter vertical profiles	*Zhao et al.* [1993]	O_3 lidar (OPAL)	10 min	$1 * 10^{-6}$ m^{-1} sr^{-1}	30% aerosol backscatter
BL wind/aerosol/turbulence	*Grund et al.* [2001]	Doppler lidar (HRDL)	0.5 s	2–6 km	10–12 cm s^{-1}
Wind/temperature profiles	*Law et al.* [2002]	915 MHz wind profiler	5 min	0.5–5 km	±1.4 ms^{-1}
Temp/RH profiles	*Wolfe et al.* [2006]	sondes	5 s	0.1–18 km	±0.3 C ±4%
LWP	*Zuidema et al.* [2005]	microwave radiometer	5 s	20 gm^{-2}	±10%
Cloud height	*Fairall et al.* [1997]	Ceilometer	15 s	0.1–7.5 km	±30 m
Cloud drop size, updraft velocity	*Kollias et al.* [2001]	3 mm Doppler radar	5 s	0.2–12 km	. . .
Turbulent fluxes	*Fairall et al.* [2003, 2006]	bow-mounted EC flux package	20 Hz, 10 min, 1 hour	2 Wm^{-2}, 0.002 Nm^{-2}	±25% at 1 hour
Low altitude temperature profiles	*Cimini et al.* [2003]	60 GHz scanning microwave radiometer	10 s	0–0.5 km	±0.3°C
Wind profiles/ microturbulence below cloud	*Frisch et al.* [1989] and *Comstock et al.* [2005]	C-band radar	5 min	0.1–2 km	±1.0 ms^{-1}

the plumes in real time. Two types of balloons were flown: NOAA's SMART balloons measured meteorological parameters, sea surface temperatures, and ozone over the Gulf of Maine and North Atlantic with one flight reaching Europe. Smaller Controlled Meteorological (CMET) balloons measured primarily winds and temperatures, but were able to be vehicle launched from multiple locations in order to target specific plumes. Version 4.1 of the Smart Balloon was employed for the ICARTT flight series [*Businger et al.*, 2006]. Previous versions of the balloon and its deployment in field campaigns have been described by *Johnson et al.* [2000] and *Businger et al.* [1999]. The balloons were released from the town of Orient on the promontory tip of the northern peninsula of Long Island, New York. Four balloons were released with flight durations over the North Atlantic ranging from 2 to 12.3 days and travel distances of 1,030 to 6,780 km. Five CMET balloons tracked urban air pollution plumes over New England and the Gulf of Maine, eastern Canada, and the Atlantic Ocean. They were vehicle launched into emerging urban plumes from New York and Boston. Flights ranging from 12 to 120 hours in duration measured the quasi-Lagrangian evolution of the low-level winds, temperature, and, in one case, ozone and relative humidity, downwind of major source regions.

[90] Two additional aircraft, the NASA DC-8 and the NASA J-31, are described by the *Singh et al.* [2006] overview paper in the INTEX-A/ICARTT special section in *Journal of Geophysical Research*.

Appendix B: Surface Site Instrumentation and Other Details

[91] The Chebogue Point site (43.75°N, 66.12°W) was instrumented to study outflow of air pollution from North America with a focus on aerosol composition and ozone photochemistry. Chebogue Point is located at the southwest tip of Nova Scotia (Figure B1), 9 km

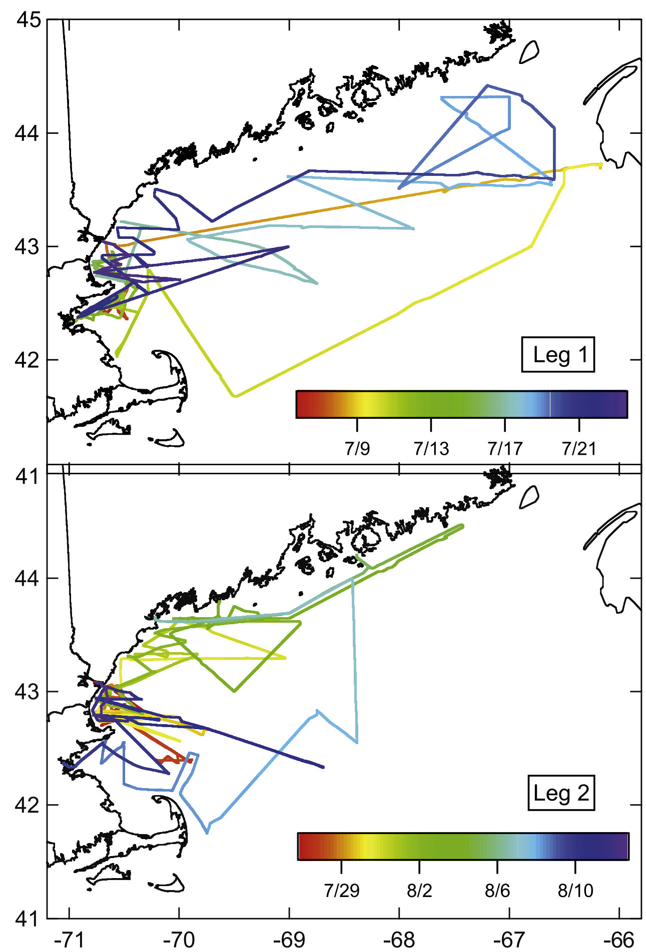

Figure A3. Cruise tracks of NOAA R/V *Ronald H. Brown* during ICARTT.

Table A4a. FAAM BAE146-301 Aircraft Instrumentation for Gas-Phase Measurements

Species/Parameter	Reference	Technique	Averaging Time	Accuracy	Precision	Detection Limit
NO	*Brough et al.* [2003]	NO/O_3 chemiluminescence	1 s	10 s	12%	40 ppt
NO_2	*Brough et al.* [2003]	photolysis-chemiluminescence	1 s	10 s	35%	350 ppt
NO_y	*Brough et al.* [2003]	Au converter-chemiluminescence	1 s	10 s	21%	70 ppt
O_3		UV absorption	3 s	5%	1 ppbv	2 ppbv
CO	*Gerbig et al.* [1999]	VUV resonance fluorescence	1 s		1 ppbv	2 ppbv
H_2O	. . .	Lyman alpha absorption and dew point	1 s	±1°	. . .	. . .
NMHCs (C_2–C_8), DMS, acetone	*Schauffler et al.* [1999]	grab sample/GC	60 s	5–10%	1–3%	10–1 pptv
Halocarbons (C_1–C_2)	*Schauffler et al.* [1999]	grab sample/GC	60 s	5–10%	1–5%	0.1 ppt
Alkylnitrates (C_1–C_5)	*Schauffler et al.* [1999]	grab sample/GC	60 s	5–20%	1–5%	0.005 ppt
VOCs	. . .	proton transfer reaction mass spectrometer	1–2 s	10–50%	10%	20–80 ppt
PAN	*Roberts et al.* [2004]	dual GC/ECD	≈90 s	10%	3%	10 pptv
HCHO	*Cárdenas et al.* [2000]	Hantzsch fluorometric	10 s	30%	12%	50 pptv
Peroxides (inorganic and organic)	*Penkett et al.* [1995]	fluorometric	10 s			5 pptv
Peroxy radicals (RO_2 + HO_2)	*Green et al.* [2006] and *Monks et al.* [1998]	chemical amplifier	30–60 s	±40%	6%	2 pptv

Table A4b. FAAM BAE146-301 Aircraft Instrumentation for Aerosol and Ancillary Data Measurements

Species/Parameter	Reference	Technique	Averaging Time	Detection Limit
Position, winds, u,v,w	. . .	INS, GPS, 5 port turbulence probe	0.1 s	~0.01 $\Delta p/P_s$
Black carbon		particle soot absorption photometer		
CCN	*Stolzenburg and McMurry* [1991]	condensation particle counter	1 s	0 cm^{-3}
Aerosol bulk composition	*Jayne et al.* [2000]aerosol mass spectrometer (AMS)	30 s	15–150 ng m^{-3} (species-dependent)	
NO_2 photolysis $j(NO_2$	*Junkerman et al.* [1989] and *Volz-Thomas et al.* [1996]	fixed bandwidth radiometry	1 s	. . .
O_3 photolysis $j(O^1D)$	*Junkerman et al.* [1989] and *Volz-Thomas et al.* [1996]	fixed bandwidth radiometry	1 s	. . .

Table A5. FAAM BAE146-301 Flights

Flight	Flight Description	Date in 2004	Takeoff-Landing, UT
B028	transit Cranfield, U.K., to Faial, Azores (refuel Oporto); fire plumes encountered in U.K. SW approaches	12 Jul	0930–2130
B029	northwest of Azores, low-level U.S. outflow and Alaskan fires	15 Jul	0842–1326
B030	south and west of Azores, low/midlevel polluted features from United States	17 Jul	1256–1737
B031	north of Azores to aircraft range limit into New York plume	19 Jul	0904–1405
B032	major midtroposphere interception of biomass burning plumes	20 Jul	0837–1315
B033	to west of Azores for ENVISAT underpass and low-level pollution	22 Jul	0920–1349
B034	reinterception of New York plume and outflow from Africa, refuel Santa Maria	25 Jul	0928–1624
B035	DC8 intercomparison to west of the Azores mainly in clean marine air	28 Jul	1157–1632
B036	upper level export in WCB from U.S. + Alaskan fires at higher T	29 Jul	0830–1300
B037	low-level export ahead of cold front sampled by P3, + fires + stratosphere influence	31 Jul	0830–1315
B038	north of Azores, targeting same air mass ahead of cold front	1 Aug	0744–1244
B039	transit Faial, Azores, to Cranfield, U.K. (refuel Oporto), with DLR Falcon intercomparison over Brittany, France	3 Aug	0722–1514

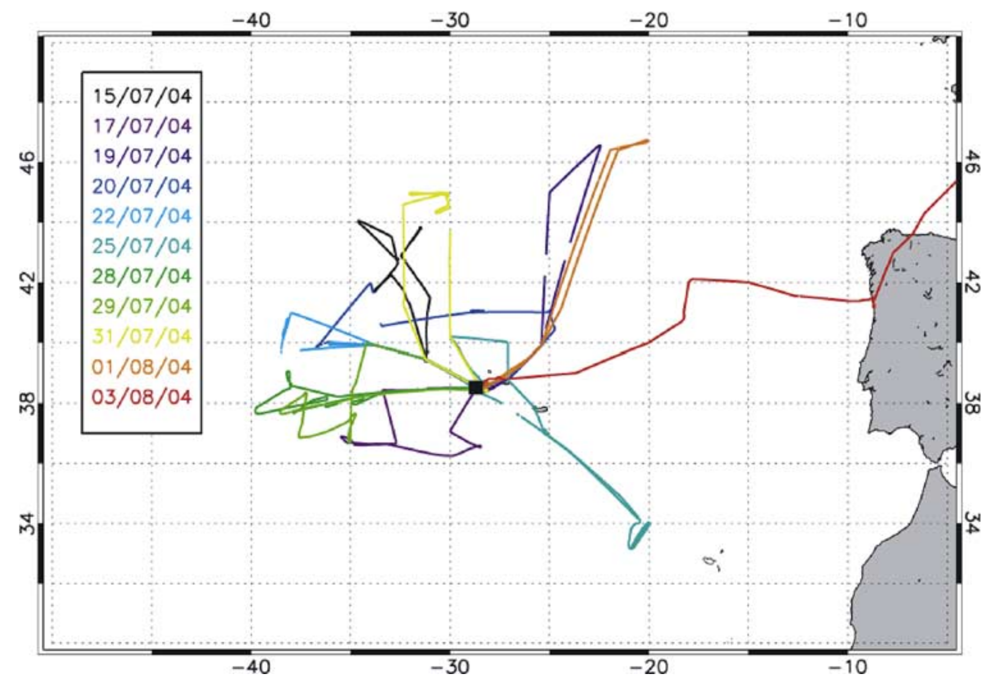

Figure A4. Map indicating FAAM BAE146 aircraft flights during ICARTT.

Table A6a. DLR Falcon Gas-Phase and Ancillary Measurements

Species/Parameter	Reference	Technique	Averaging Time	Accuracy	Precision	Detection Limit
NO	*Schlager et al.* [1997]	NO/O_3 chemiluminescence	1 s	7%	3%	2 pptv
NO_y	*Ziereis et al.* [2000]	Au converter-chemiluminescence	1 s	12%	5%	15 pptv
O_3	*Schlager et al.* [1997]	UV absorption	5 s	5%	2%	0.5 ppbv
CO	*Gerbig et al.* [1996]	VUV resonance fluorescence	5 s	5%	2%	1 ppbv
CO	*Wienhold et al.* [1998] and *Fischer et al.* [2002]	TD-LAS	5 s	7%	3%	2 ppbv
NMHCs (C_2-C_{10})	*Rappenglück et al.* [1998]	grab sample/GC	60 s	5–10%	1–5%	3 pptv
CH_4	*Wienhold et al.* [1998]	TD-LAS	5 s	7%	5%	0.03 ppmv
CO_2	*Fischer et al.* [2002]	IR-absorption	1 s	2%	0.1%	0.3 ppbv
SO_2	*Speidel et al.* [2006]	ion trap mass spectrometry	2 s	10%	3%	10 pptv
$J(NO_2)$	*Volz-Thomas et al.* [1996]	filter radiometry	1 s	5E-4 s^{-1}	1E-4 s^{-1}	. . .
Humidity	*Schumann et al.* [1995]	Lyman alpha absorption	1 s	0.3 g m^{-3}	0.01 g m^{-3}	. . .
Temperature	*Schumann et al.* [1995]	Pt 100, Pt 500	1 s	0.5°	0.1°	. . .
Wind (horizontal, vertical)	*Schumann et al.* [1995]	INS, GPS, five hole probe	1 s	1 m s^{-1} (horizontal), 0.3 m s^{-1} (vertical)	0.1 m s^{-1} (horizontal), 0.05 m s^{-1} (vertical)	. . .

Table A6b. DLR Falcon Aircraft Instrumentation for Aerosol Measurements

Species/Parameter	Reference	Technique	Averaging Time	Detection Limit
Ultrafine particle size distribution	*Schröder and Ström* [1997] and *Feldpausch et al.* [2006]	condensation particle counters operated at different lower cutoff diameters and diffusion screen separator	5 s	1 cm^{-3}
Aitken mode size distribution	A. Petzold et al. (unpublished manuscript, 2006)	differential mobility analyser (DMA)	70 s	1 cm^{-3}
Accumulation mode size distribution (dry state)	*Petzold et al.* [2002]	passive cavity aerosol spectrometer probe (PCASP 100X)	5 s	0.1 $cm^{-3?}$
Volume fraction of volatile/refractory particles	*Clarke* [1991]	thermodenuder connected to condensation particle counters	5 s	
Volume absorption coefficient	*Bond et al.* [1999]	particle soot absorption photometer (PSAP)	20 s	0.1 Mm^{-1} at STP

Table A7. DLR Falcon Flights[a]

Flight	Flight Description	Date in 2004	Takeoff-Landing, UT
1	Oberpfaffenofen (near Munich) to Po valley: NA BB and urban plume	2 Jul	1314–1519
2	Cranfield to North Sea: intercomparison with BAe146	7 Jul	1150–1411
3	Oberpfaffenhofen to Po valley: urban plume	13 Jul	0805–1050
4	Transfer Oberpfaffenhofen to Creil (near Paris)	19 Jul	0923–1047
5	Creil to San Sebastian (Sp): New York/Boston plume, NA BB plume	22 Jul	0940–1057
6	San Sebastian to Creil: New York/Boston plume, NA BB plume	22 Jul	1505–1703
7	Creil to Brest to English Channel: NA BB plume, ship emissions	23 Jul	1211–1602
8	Creil to Shannon (Ireland): New York/Boston plume, NA BB plume	25 Jul	1337–1640
9	Shannon to Creil: New York Boston plume, NA BB plume	25 Jul	1742–1953
10	Creil to English Channel: New York/Boston plume, London plume	26 Jul	1507–1850
11	Creil to Gulf of Biscay: NA BB plume, ship emissions	30 Jul	1500–1835
12	Creil to northern France: upper level outflow from USA	31 Jul	1207–1355
13	Creil to northwest France: intercomparison Falcon with BAe146	3 Aug	1424–1725

[a]BB, biomass burning; NA, North America.

south-southwest of Yarmouth, 130 km southeast of the Maine coastline, 430 km northeast from Boston, Massachusetts, and 730 km northeast from New York, New York. Measurements included a broad array of trace gas, aerosol, radiation, and meteorological measurements (Tables B1a and B1b). Most of the sampling inlets were mounted on a 10 m scaffolding tower, and instruments were housed in climate-controlled laboratories at the base of the tower. The site operated continuously from 1 July through 15 August 2004.

[92] The radar wind profiler network comprised ten land-based and one shipboard 915-MHz Doppler radar

Table A8. Twin Otter Aircraft Instrumentation for Aerosol and Ancillary Data Measurements

Parameter	Reference	Technique	Averaging Time	Detection Limit	Size Range Detected
Particle number concentration	*Mertes et al.* [1995] and *Buzorius* [2001]	condensation particle counter (TSI CPC 3010)	1 s	0–10,000 particles/cm^{-3}	$D_p > 10$ nm
Cloud condensation nuclei concentration	*Rissman et al.* [2006]	linear temperature gradient growth chamber with optical detection (Caltech three-column CCN counter)	1 s	0–10,000 particles/cm^{-3}	N/A
Aerosol size distributions at dry and humid condition	*Wang and Flagan* [1990] and *Wang et al.* [2003]	scanning differential mobility analyzer (dual automated classified aerosol detector (DACAD))	73 s	N/A	10–700 nm
Aerosol size distribution		passive cavity aerosol spectrometer probe (PCASP)	1 s	N/A	0.1–2.6 μm
Aerosol bulk ionic composition and soluble organic composition	*Weber et al.* [2001] and *Sorooshian et al.* [2006a]	particle-into-liquid sampler (PILS)	5 m	0.02–0.28 μg/m^3 (depending on species)	<1 μm
Aerosol bulk composition (nonrefractory species)	*Jayne et al.* [2000] and *Bahreini et al.* [2003]	Aerodyne quadrupole aerosol mass spectrometer (AMS)	30 s or 1 m	0.2–2.3 μg/m^3 (depending on species)	$D_{va} \sim 40$ nm to 1 μm
Aerosol organic functional group	Gilardoni et al. (submitted manuscript, 2006)	FTIR spectroscopy of <1 μm particles on Teflon filters	~1 hour	N/A	<1 μm
Soot absorption	*Arnott et al.* [1999, 2006]	photoacoustic absorption spectrometer	1 s	1 Mmn^{-1}	10 nm to 5 μm
Soot absorption	*Bond et al.* [1999]	particle soot absorption photometer (PSAP)	1 s or higher	N/A	N/A
Soot absorption	*Baumgardner et al.* [2004]	single particle soot photometer (SP2)	N/A	N/A	150 nm to 1.5 μm
Separation of cloud droplets from interstitial aerosol	*Noone et al.* [1988]	counterflow virtual impactor	N/A	N/A	N/A
Cloud droplet size distribution	*Baumgardner et al.* [2001]	cloud, aerosol, and precipitation spectrometer (CAPS)	1 s	0–1,000 particles/cm^{-3}	0.4 μm to 1.6 mm
Cloud droplet size distribution	*Cerni* [1983]	forward scattering spectrometer probe (FSSP)	1 s	N/A	1–46 μm
Cloud droplet liquid water content	*Gerber et al.* [1994]	light diffraction (Gerber PVM-100 probe)	1 s	N/A	~5–50 μm

Table A9. CIRPAS Twin Otter Flights

Flight	Flight Description	Date in 2004	Takeoff/Landing, UTC
1	aerosol characterization over NW Ohio and Indiana, Convair coordination	2 Aug	1507–2032
2	clouds south of Cleveland	3 Aug	1657–2152
3	Conesville power plant plume and cloud, Convair coordination	6 Aug	1617–2041
4	Conesville power plant plume in clear air	8 Aug	1818–2145
5	Conesville power plant plume and cloud	9 Aug	1709–2216
6	Monroe power plant plume and cloud	10 Aug	1804–2300
7	cloud physics at SE shore of Lake Erie	11 Aug	1754–2246
8	pollution from Detroit, Monroe power plant plume, Convair coordination	13 Aug	1831–2303
9	cloud physics SW of Cleveland, Convair coordination	16 Aug	1816–2237
10	cloud physics SW of Cleveland, Convair coordination	17 Aug	1813–2124
11	clouds, SW of Ontario, Convair coordination	18 Aug	1537–1910
12	Conesville power plant plume and cloud	21 Aug	1740–2252

wind profilers [*Carter et al.*, 1995] that measured winds in the planetary boundary layer (see Figure B1 and Table B2). Typical vertical coverage was from 120 m to ~4000 m above the surface, depending on atmospheric conditions. Radio acoustic sounding systems (RASS) were operated in conjunction with most of the wind profilers to measure temperature profiles up to ~1500. The vertical resolutions of both the wind and temperature measurements were either 60 m or 100 m. The wind profiler data were quality controlled after the data collection period using the continuity technique developed by *Weber et al.* [1993].

[93] Operation of the wind profiler on the R/V *Ronald H. Brown* was hindered by sea clutter (i.e., sidelobe reflections from the ocean surface), which often prevented wind retrievals in approximately the lowest 500 m above the surface. A Doppler lidar on the *Ronald H. Brown* measured winds below clouds with up to 5 m resolution using the velocity-azimuth display (VAD) technique [*Browning and Wexler*, 1968]. After the study, the lidar wind profiles were merged with wind profiler data to take advantage of the unique measurement capabilities of each instrument [*Wolfe et al.*, 2006].

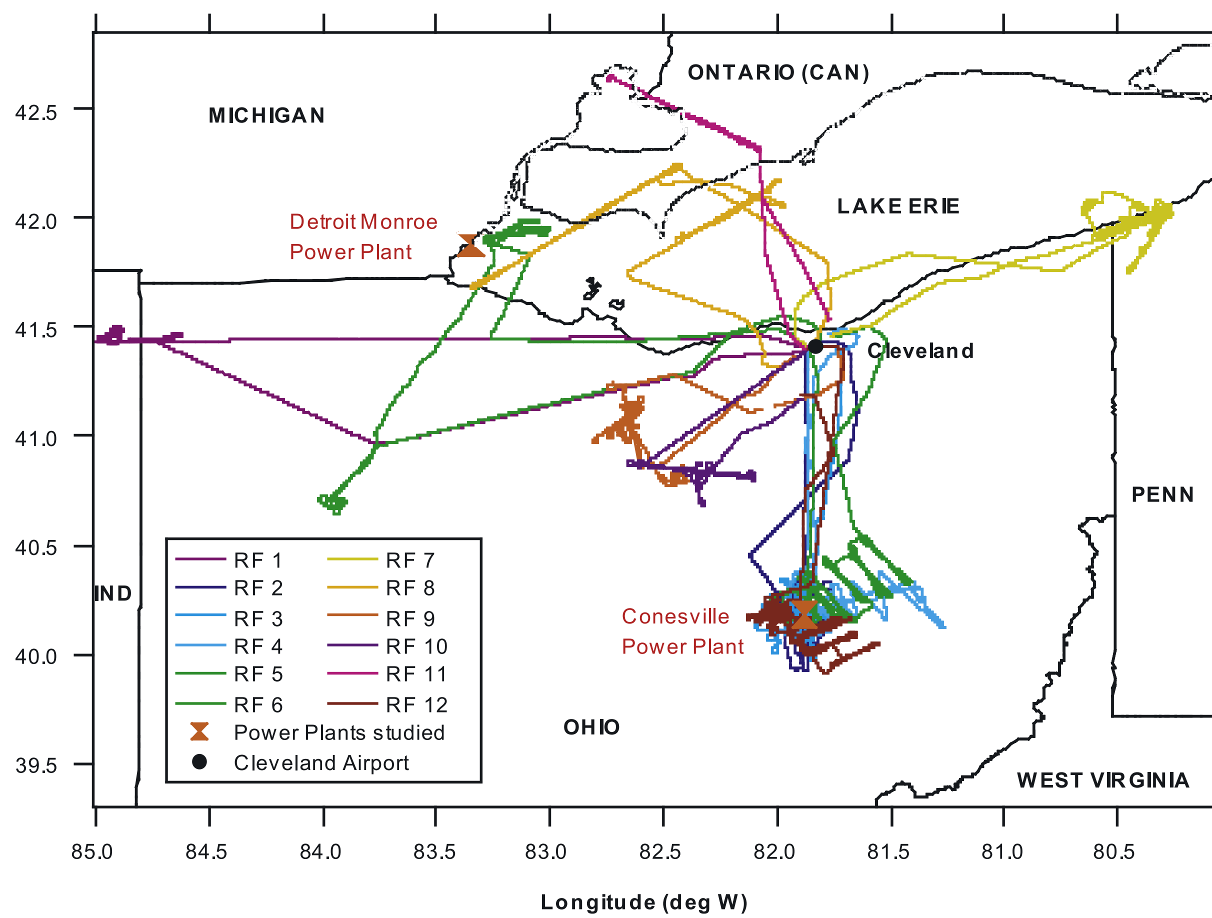

Figure A5. Flight tracks of the CIRPAS Twin Otter during ICARTT.

Table A10. Canadian Convair 580 Flights

Flight	Flight Description	Date in 2004	Takeoff-Landing, UT
1	out of Cleveland to 20,000′ over Lake Erie and in BL southeast of Cleveland	21 Jul	1754–2013
2	transit, Cleveland to Bangor, Maine, for TIMs	21 Jul	2213–0053
3	TIMs flight from Bangor with profiles over Fundy and at Chebogue Point	22 Jul	1524–1913
4	TIMs flight from Bangor with profiles north of Saint John, Fundy and Kejimkujik	22 Jul	2035–0007
5	transit, Bangor to Cleveland	23 Jul	1524–1845
6	out of Cleveland, profile to 10000′ over Lake Erie, cloud sampling south of Lake Erie	23 Jul	2040–2354
7	evening flight to Terra Haute for aerosol nitrate, engine problem at Terra Haute	27 Jul	0014–0327
8	cloud sampling south of Cleveland	31 Jul	1801–2232
9	Cleveland to Indianapolis for forecasted aerosol nitrate	2 Aug	1201–1702
10	Indianapolis to Cleveland for nitrate, coordinated with CIRPAS Twin Otter	2 Aug	1825–2020
11	BL cloud sampling over SW Ontario	3 Aug	1457–1829
12	towering Cu sampling south of Cleveland over Ohio	3 Aug	2026–0010
13	towering Cu sampling south of Cleveland over Ohio	5 Aug	1624–2102
14	towering Cu sampling over Conesville with CIRPAS Twin Otter	6 Aug	1618–2038
15	sampling over eastern Ohio in polluted air with little cloud	9 Aug	
16	sampling aerosol and boundary layer cloud to the east and downwind of Chicago	10 Aug	1624–2015
17	sampling aerosol and cloud further east and downwind of Chicago	10 Aug	2122–0054
18	sampling boundary layer cloud over SW Ontario downwind of Detroit-Windsor	11 Aug	1829–2144
19	sampling Cumulus in boundary layer along south shore of Lake Erie	12 Aug	1740–2120
20	sampling towering Cu over Toledo and south of Akron	13 Aug	1916–2324
21	sampling moderately polluted air and clouds over Ohio	16 Aug	1846–2156
22	sampling polluted air over Ohio with little cloud	17 Aug	1802–2048
23	sampling BL cloud over SW Ontario downwind of Detroit-Windsor, coordinated with Twin Otter	18 Aug	1504–1847

[94] Figure B2 gives the locations of the AIRMAP Network sites and the CHAiOS study at the Appledore Island AIRMAP site. Tables B3a and B3b summarize the CHAiOS measurements.

[95] Atmospheric composition measurements at the PICO-NARE station in the Azores are designed to study ozone photochemistry plus aerosol absorption. Measurements began in July 2001, with CO, ozone, and black

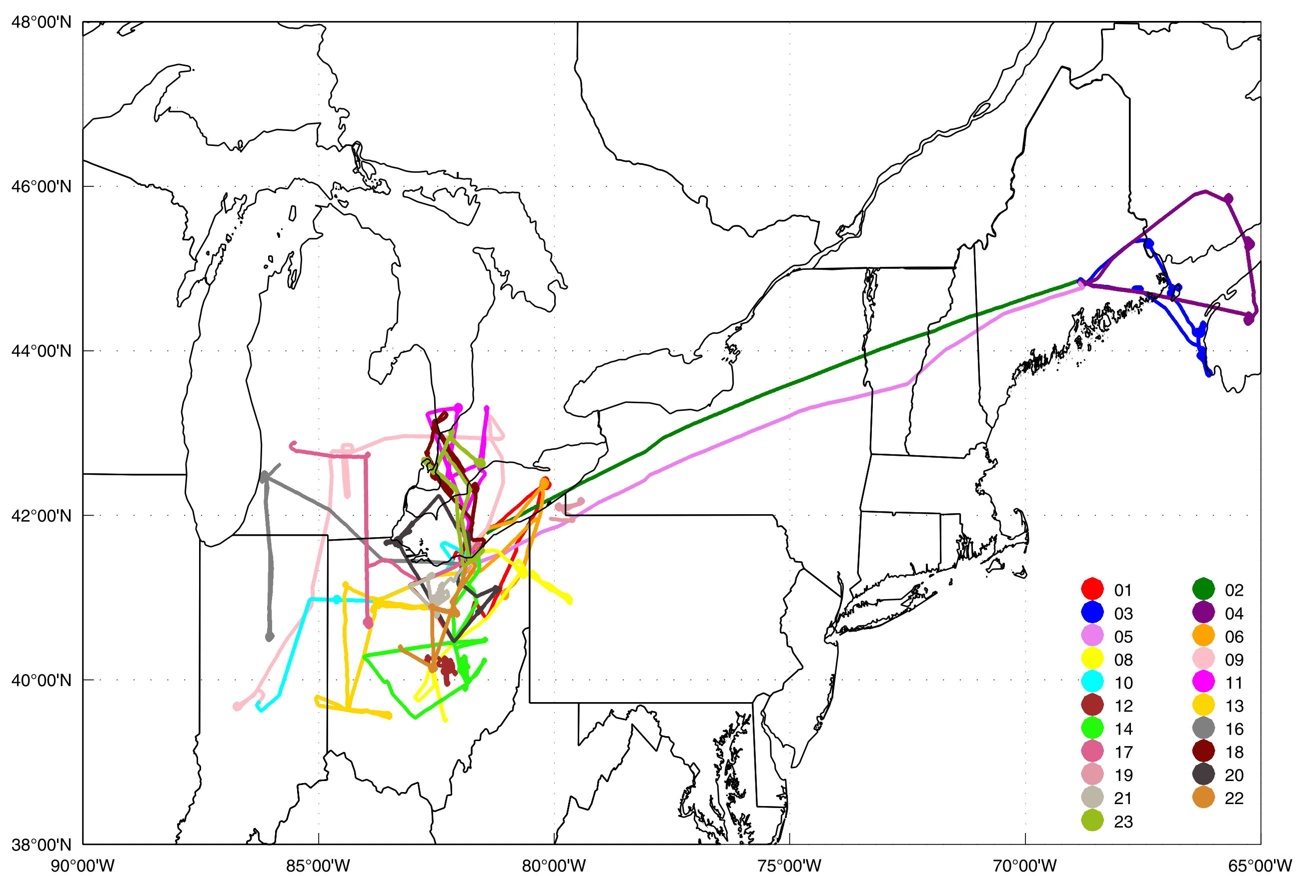

Figure A6. Flight tracks of the Canadian Convair 580 during ICARTT.

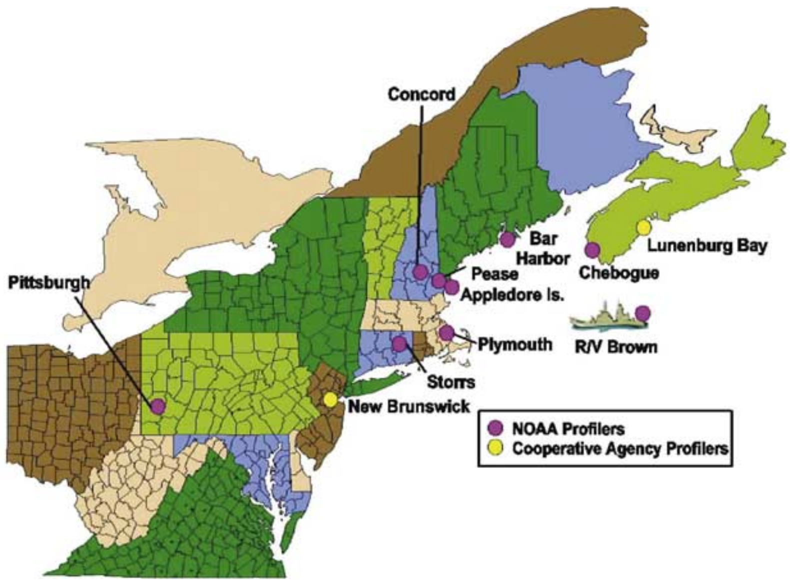

Figure B1. Map of observing sites in the ICARTT wind profiler network.

carbon. Nitrogen oxides instrumentation was added in 2002, with nearly continuous observations from spring 2004 through August 2005, and NMHC measurements began in summer 2004 and were nearly continuous fall 2004 through summer 2005. The measurement techniques are summarized in Table B4. Standard meteorological observations are also made. During the summer 2004 ICARTT period, additional meteorological stations were added along the mountainside to study upslope flow, as described by *Kleissl et al.* [2006].

[96] Eight systems contributed to the European Lidar Network during the ITOP/ICARTT experiment. Figure B3 gives a map of this network and Table B5 gives a measurement timetable. All systems measured aerosol backscatter profiles. The Observatoire de Haute Provence (OHP) and Athens systems had a UV-DIAL measurement capability and were able to provide ozone vertical profiles (respectively up to 12 km and 4 km). The joint measurements of ozone and aerosol backscatter profiles together with meteorological model simulations make possible the separation

Table B1a. Chebogue Point Instrumentation for Gas-Phase Measurements

Species/Parameter	Reference	Technique	Averaging Time	Accuracy	Precision	Detection Limit
O_3	*Goldstein et al.* [2004]	UV absorption, Dasibi 1008-RS	1 m	2%	1 ppbv	1 ppbv
CO	*Goldstein et al.* [2004]	infrared absorption, gas filter correlation, TEI 48CTL	1 m	2%	1%	20 ppbv
H_2O	*Goldstein et al.* [2004]	infrared absorption, Licor 6262	1 m	5%	1%	NA
CO_2	*Goldstein et al.* [2004]	infrared absorption, Licor 6262	1 m	1 ppm	0.2 ppm	NA
NMHCs (C_3-C_{10})	*Millet et al.* [2005, 2006]	in situ GC/MS/FID	30 m	10%	2–8%	1–25 pptv
Halocarbons (C_1-C_2)	*Millet et al.* [2005, 2006]	in situ GC/MS/FID	30 m	10%	2–7%	<1–2 pptv
Alkylnitrates (C_1-C_5)	*Millet et al.* [2005, 2006]	in situ GC/MS/FID	30 m	10–25%	9–25%	0.4–1 pptv
Oxygenated VOC (C_1-C_5)	*Millet et al.* [2005, 2006]	in situ GC/MS/FID	30 m	10–15%	4–15%	2–100 pptv
VOCs	Holzinger et al. (submitted manuscript, 2006)	PTRMS, Ionicon Analytik	1 min	10–30%	5–30%	10–250 pptv
PAN, PPN, MPAN, PiBN, APAN	M. Marchewka et al. (unpublished manuscript, 2006)	direct injection, GC/ECD	1 min, at 5 min intervals	5 pptv + 15%, 5 pptv + 20%	2%, 2%	5 pptv, 5 pptv
NO_2, ΣPNs, ΣANs, HNO_3, NO_y^*	*Day et al.* [2002]	TD-LIF	1 min	10–20%	10%	50–150 pptv
Radon	*Whittlestone and Zahorowski* [1998]	dual-flow loop, two-filter Rn detector, ANSTO Inc.	30 m	20%	8%	100 mBq m^{-3}
Total gaseous mercury	*Kellerhals et al.* [2003]	CVAFS, Tekran 2537A	5 min	2%	2%	<0.1 ng/m^3
SO_2	Aerodyne	Thermo Electron 43S SO_2 monitor	1 min		0.1 ppbv	0.1 ppbv

Table B1b. Chebogue Point Instrumentation for Aerosol and Ancillary Data Measurements

Species/Parameter	Reference	Technique	Averaging Time	Detection Limit
Aerosol mass and elemental composition	*VanCuren et al.* [2005]	8-RDI sampler and s-XRF analysis	3 hours	<ng/m^3 for elements 0.5 μg/m^3 for mass
Aerosol bulk ionic composition and total mass	*Quinn et al.* [2000]	impactors, sub-1 um and sub-10 um size fractions	12 hours	1 ng/m^3
Nonrefractory aerosol composition with aerodynamic sizing	*Jayne et al.* [2000]	Aerodyne aerosol mass spectrometer (AMS)	1 min (30 min reported)	$30 < D_{va} < 1000$ nm; 20 ng m^{-3} (SO_4^{2-}); 7 ng m^{-3} (NO_3^-); 0.17 μg m^{-3} (NH_4^+); 0.12 μg m^{-3} (OM)
Chemically resolved volatility	J. A. Huffman et al. (manuscript in preparation, 2006)	inlet thermal denuder system for AMS	20 min	as AMS
Particle optical size and density	E. S. Cross et al. (manuscript in preparation, 2006)	light scattering module for AMS	real time	$D_o > 180$ nm; $D_{va} < 1$ μm
Aerosol size distribution	*Williams et al.* [2000]	differential mobility particle sizer (DMPS)	10 min	$3 < D_{mob} < 800$ nm
Particle hygroscopic growth (D_{mob} = 40, 89 and 217 nm)	*Cubison et al.* [2005]	hygroscopicity tandem differential mobility analyzer (HTDMA)	1 hour	$0.85 < g(RH) < 2.25$
Particle volatility (D_{mob} = 130 nm)	*Burtscher et al.* [2001]	volatility tandem differential mobility analyzer (VTDMA)	15 min	N/A
Aerosol absorbance and equivalent black carbon		Thermo Electron 5012 multiangle absorption photometer (MAAP)	1 min	0.66 Mm^{-1} (B_{abs}); 0.1 μg m^{-3} (BC)
Particle number concentrations		TSI 3022a condensation particle counter (CPC)	1 min	$D > 7$ nm
Particle number concentration	*Hering et al.* [2005]	water condensation particle counter (Quant 400, prototype for TSI 3785)	1 min	5 nm
Aerosol organic functional groups	Gilardoni et al. (submitted manuscript, 2006)	FTIR spectroscopy of <1 μm particles on Teflon filters	4–12 hours	1 μg accuracy 15%
Speciated organic composition	*B. J. Williams et al.* [2006]	thermal desorption aerosol GC/MS/FID (TAG)	30 min	typically 0.05–0.7 ng/cm^3
Wind profiles	*White et al.* [2006b]	915-MHz radar wind profiler	60 min	
Temperature profiles	*White et al.* [2006b]	radio acoustic sounding system	5 min	
Cloud and aerosol backscatter	Duck et al. (submitted manuscript, 2006)	lidar	1 min	
Aerosol optical depth and size distribution	Duck et al. (submitted manuscript, 2006)	Sun photometer	1 min	
Aerosol number concentration	*Sinclair and Hoopes* [1975] and *Delene and Ogren* [2002]	TSI 3010 condensation particle counter (CPC)	1 min	0/cm^3
Aerosol light absorption	*Bond et al.* [1999] and *Delene and Ogren* [2002]	Radiance Research particle soot absorption photometer (PSAP)	1 min	0.9 Mm^{-1} noise
Aerosol total and back light scattering at three wavelengths	*Ahlquist and Charlson* [1967] and *Delene and Ogren* [2002]	TSI 3563 integrating nephelometer	1 min	1.8 Mm^{-1} noise at 20 Mm^{-1} scattering
Hygroscopic growth (f(RH))	*Rood et al.* [1989]	humidograph (humidity conditioner plus second TSI 3563 nephelometer)	30 min	
Cloud condensation nuclei (CCN) at five supersaturations	*Roberts and Nenes* [2005]	Droplet Measurement Technologies CCN counter	30 min	~0.75 um lowest size bin
Aerosol size distribution (0.02–0.5 μm)	*Buzorius et al.* [2004]	scanning electrical mobility sizer, Brechtel Manufacturing Inc.	1.2 min	0.01 μm to 20 cm^{-3} μm^{-1}, 0.5 μm → 4 cm^{-3} μm^{-1}
Wind speed and direction	*Goldstein et al.* [2004]	propeller anemometer, R.M. Young	30 min	
RH, Tair	*Goldstein et al.* [2004]	Vaisala Inc., model HMP45C	30 min	
Photosynthetically active radiation	*Goldstein et al.* [2004]	Quantum Sesnor, LiCor Inc., 190SZ	30 min	

Table B2. Locations of NOAA and Cooperative Agency Boundary Layer Wind Profilers Available for the ICARTT Study

Location	Designation	Latitude	Longitude	Elevation	RASS	Sponsor
Appledore Island, Maine	ADI	42.99	−70.62	5 m	yes	NOAA
Bar Harbor, Maine	BHB	44.44	−68.36	4 m	yes	NOAA
Chebogue Pt., Nova Scotia	CHE	43.70	−66.10	15 m	yes	NOAA
Concord, New Hampshire	CCD	43.21	−71.52	104 m	yes	NOAA
Lunenburg Bay, Nova Scotia	LUN	44.40	−64.30	30 m	yes	Environment Canada
New Brunswick, New Jersey	RUT	40.50	−74.45	10 m	yes	Rutgers University and New Jersey Department of Environmental Protection
Pease International Tradeport, New Hampshire	PSE	43.09	−70.83	30 m	yes	NOAA
Pittsburgh, Pennsylvania	PIT	40.48	−80.26	335 m	yes	NOAA
Plymouth, Massachusetts	PYM	41.91	−70.73	46 m	yes	NOAA
R/V *Ronald H. Brown*	RHB	variable	variable	5 m	no	NOAA
Storrs, Connecticut	STS	41.80	−72.23	198 m	no	NOAA

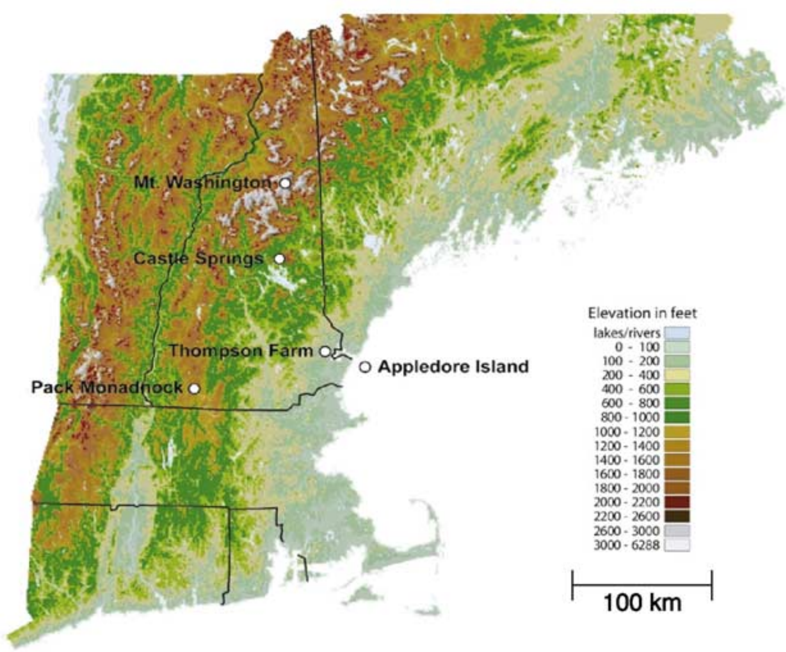

Figure B2. Map of AIRMAP observational network.

Table B3a. CHAiOS Instrumentation for Gas-Phase Measurements

Species/Parameter	Reference	Technique	Averaging Time	Accuracy	Precision	Detection Limit
NO_2	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	0.15 or 0.05 ppbv	greater of 3% or accuracy	accuracy × 2
HCHO	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	0.3 ppbv	greater of 3% or accuracy	accuracy × 2
O_3	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	2 ppbv	greater of 3% or accuracy	accuracy × 2
HONO	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	0.05 ppbv	greater of 3% or accuracy	accuracy × 2
NO_3	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	1.7 ppbv	greater of 3% or accuracy	accuracy × 2
SO_2	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	0.07 ppbv	greater of 3% or accuracy	accuracy × 2
BrO	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	~0.6 pptv	greater of 3% or accuracy	accuracy × 2
OIO	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	1 to 5 pptv	greater of 3% or accuracy	accuracy × 2
IO	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	0.6 pptv	greater of 3% or accuracy	accuracy × 2
I_2	*Alicke et al.* [2002] and *Stutz et al.* [2002]	long-path DOAS	5 to15 m	4 to 25 pptv	greater of 3% or accuracy	accuracy × 2
NO_2	Pikelnaya et al. (submitted manuscript, 2006)	MAX-DOAS	5 to15 m	5.0×10^{14} molec cm^{-2}	greater of 3% or accuracy	accuracy × 2
HCHO	Pikelnaya et al. (submitted manuscript, 2006)	MAX-DOAS	5 to15 m	1.1×10^{16} molec cm	greater of 3% or accuracy	accuracy × 2
BrO	Pikelnaya et al. (submitted manuscript, 2006)	MAX-DOAS	5 to15 m	1×10^{13} molec cm	greater of 3% or accuracy	accuracy × 2
OIO	Pikelnaya et al. (submitted manuscript, 2006)	MAX-DOAS	5 to15 m	1.5×10^{13} molec cm	greater of 3% or accuracy	accuracy × 2
IO	Pikelnaya et al. (submitted manuscript, 2006)	MAX-DOAS	5 to15 m	5×10^{12} molec cm	greater of 3% or accuracy	accuracy × 2
I_2	Pikelnaya et al. (submitted manuscript, 2006)	MAX-DOAS	5 to15 m	5×10^{13} molec cm	greater of 3% or accuracy	accuracy × 2
HCOOH	*Pszenny et al.* [2004]	tandem mist chamber	2 hours	~15%	greater of 10–15% or 0.5 × DL	5 pptv
CH_3COOH	*Pszenny et al.* [2004]	tandem mist chamber	2 hours	~15%	greater of 10–15% or 0.5 × DL	3 pptv
HCl	*Pszenny et al.* [2004]	tandem mist chamber	2 hours	~15%	greater of 10–15% or 0.5 × DL	10 pptv
HNO_3	*Pszenny et al.* [2004]	tandem mist chamber	2 hours	~15%	greater of 10–15% or 0.5 × DL	11 pptv
NH_3	*Pszenny et al.* [2004]	tandem mist chamber	2 hours	~15%	greater of 10–15% or 0.5 × DL	8 pptv
Cl[a]	*Pszenny et al.* [2004]	tandem mist chamber	2 hours	~15%	greater of 15% or 10 pptv	5 pptv
Total inorganic Br	*Rahn et al.* [1976]	filter pack	15 hours (daytime) or 9 hours (nighttime)	~15%	22%	0.06 pptv
Total inorganic I	*Rahn et al.* [1976]	filter pack	15 hours (daytime) or 9 hours (nighttime)	~15%	10%	0.25 pptv
$C_2–C_{10}$ NMHCs	*Zhou et al.* [2005]	canisters	5 m (hourly)	5%	0.1–3% ($C_2–C_5$); 5% ($C_6–C_{10}$)	2 pptv ($C_2–C_5$); 3 pptv ($C_6–C_{10}$)
$C_2–C_{10}$ NMHCs	*Talbot et al.* [2005]	PTR-MS	10 m	10%	15%	10–100 pptv
$C_2–C_{10}$ NMHCs	*Sive et al.* [2005]	GC-FID/ECD/MS	7.5 m every 40 m	5%	0.3–3% ($C_2–C_5$); 5–7% ($C_6–C_{10}$)	2 pptv ($C_2–C_5$); 3 pptv ($C_6–C_{10}$)
Halocarbons	*Zhou et al.* [2005]	canisters	5 m (hourly)	5–20%	1–13%	CH_3Cl, 50 pptv; CH_3Br, CH_3I, 1 pptv; C_2H_5I, 0.001 pptv; others, 0.01 pptv
Halocarbons	*Sive et al.* [2005]	GC-FID/ECD/MS	7.5 m every 40 m	5–20%	1–13%	CH_3Cl, 25 pptv; CH_3Br, CH_2Cl_2, 1 pptv; C_2H_5I, 0.001 pptv; others, 0.01 pptv
$C_1–C_5$ alkyl nitrates	*Zhou et al.* [2005]	canisters	5 m (hourly)	10%	5%	0.01 pptv
Alkyl nitrates	*Sive et al.* [2005]	GC-FID/ECD/MS				
OCS	*Zhou et al.* [2005]	canisters	5 m (hourly)	10%	4%	50 pptv
OCS	*Sive et al.* [2005]	GC-FID/ECD/MS	7.5 m every 40 m	5–20%	1–13%	25 pptv
OVOCs	*Talbot et al.* [2005]	PTR-MS	10 m	10%	15%	10–100 pptv
OVOCs	*Sive et al.* [2005]	GC-FID/ECD/MS	7.5 m every 40 m	10%	5–10%	10–100 pptv

[a]HOCl, Cl_2 and other inorganic Cl gases besides HCl.

Table B3b. CHAiOS Instrumentation for Aerosol and Ancillary Data Measurements

Species/Parameter	Reference	Technique	Averaging Time	Detection Limit
Aerosol bulk and size-segregated ionic composition	*Pszenny et al.* [2004]	bulk filters and cascade impactors/ion chromatography	15 hours (daytime) or 9 hours (nighttime)	~0.2 to ~50 ng m^{-3} for individual species
Aerosol total Br and I	*Pszenny et al.* [2004] and *Rahn et al.* [1976]	bulk filters and cascade impactors/neutron activation	15 hours (daytime) or 9 hours (nighttime)	Br ~ 0.2 ng m^{-3}, I ~ 1.5 ng m^{-3}
Aerosol organic functional groups	Gilardoni et al. (submitted manuscript, 2006)	FTIR spectroscopy of <1 μm particles on Teflon filters	4–12 hours	1 μg
Aerosol number	Russell et al. (submitted manuscript, 2006)	CNC (TSI 3025)	1 s	~5 cm^{-3}
Aerosol size distribution	Russell et al. (submitted manuscript, 2006)	DMA and APS	3 m	. . .
Photolytic flux		Bentham spectroradiometer, model DMc 150 FC	5 m	unknown

Table B4. PICO-NARE Instrumentation

Species/Parameter	Reference	Technique	Averaging Time	Accuracy (2-sigma)	Precision (2-sigma)	Detection Limit	Measurement Period
NO	*Ryerson et al.* [1999] and *Val Martín et al.* [2006]	NO/O_3 chemiluminescence	30 s	4% + 1.5 pptv	8 pptv	5 to 6 pptv (1-hour average)	2002–2005
NO_2	*Ryerson et al.* [1999] and *Val Martín et al.* [2006]	photolysis-chemiluminescence	30 s	4% + 4 pptv	17 pptv	11 to 13 pptv (1-hour average)	2002–2005
NO_y	*Ryerson et al.* [1999] and *Val Martín et al.* [2006]	Au converter-chemiluminescence	20 s	+8/−15% + 2 pptv	11 pptv	7 to 9 pptv (1-hour average)	2002–2005
O_3	*Ryerson et al.* [1998], *Honrath et al.* [2004], and *Owen et al.* [2006]	ultraviolet absorption	60 s	3%	<1 ppbv	1 ppbv	2001–2005
CO	*Honrath et al.* [2004] and *Owen et al.* [2006]	nondispersive infrared absorption	30 min	7%	4 to 9 ppbv	2 ppbv	2001–2005
Equivalent black carbon	*Fialho et al.* [2005]	multiwavelength aethalometer	1 hour	not characterized	25 ng/m^3	25 ng/m^3	2001–2005
NMHCs (C_2–C_6)	*Tanner et al.* [2006]	continuous GC	12 min and 60 min	5–10%	5–10%	<10 pptv	2004–2005

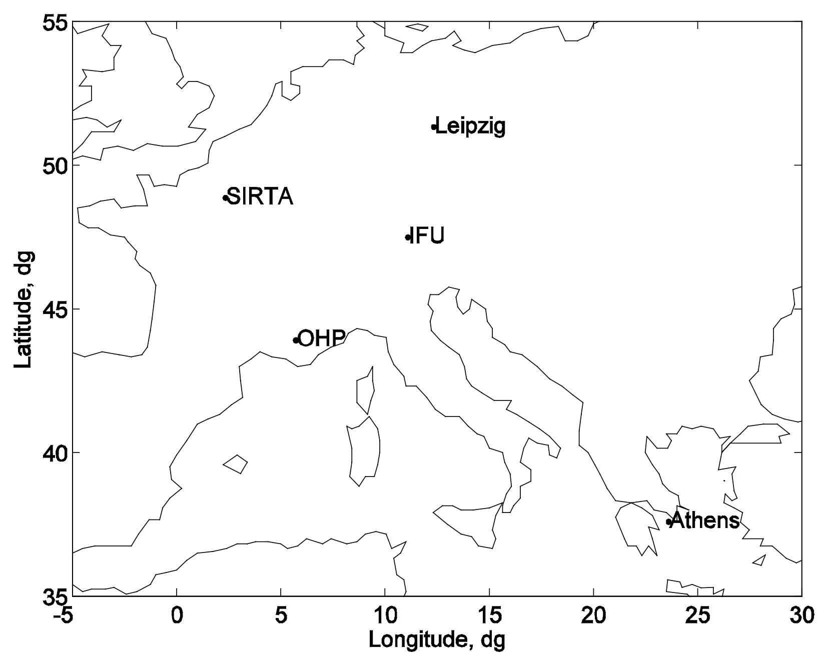

Figure B3. Map of sites in the European Lidar Network.

Table B5. Times of Measurements by European Lidar Network[a]

	5 Jul	19 Jul	20 Jul	21 Jul	22 Jul	23 Jul	24 Jul	25 Jul	26 Jul	27 Jul	28 Jul	29 Jul	30 Jul	31 Jul	1 Aug	2 Aug	3 Aug
OHP O_3, aerosol		1500–2100	1500–2000	0700–1700	0800, 1900	1900	0700–2000	1800–2000	0600–2400	0000–2000	0800, 2000	0500–2200	0500–2200	0500–2200	1800–2100	0600, 2000	0600
SIRTA aerosol	0600–1800	0600–1700	0700		0700	0500–1200			0600–1500	0600–1800	0600–1800	0600–1800	0600–1600			0700–1800	0600
Athens O_3				0800, 1500							1300						0600–1200
Athens aerosol	1200	0800–2100		1100					0800–2100							2100	
IFU aerosol			x	x		x					x	x					
Leipzig aerosol	x	x			x							x					x
Hamburg aerosol		x			1300–2100											x	
Potenza aerosol		1300–2100	0900–2100		1700–2100				2000–2200			1600–2100				0800–2400	1200–2200

[a]Times are in UT.

between export of European pollution above the PBL and layers related to long-range transport.

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