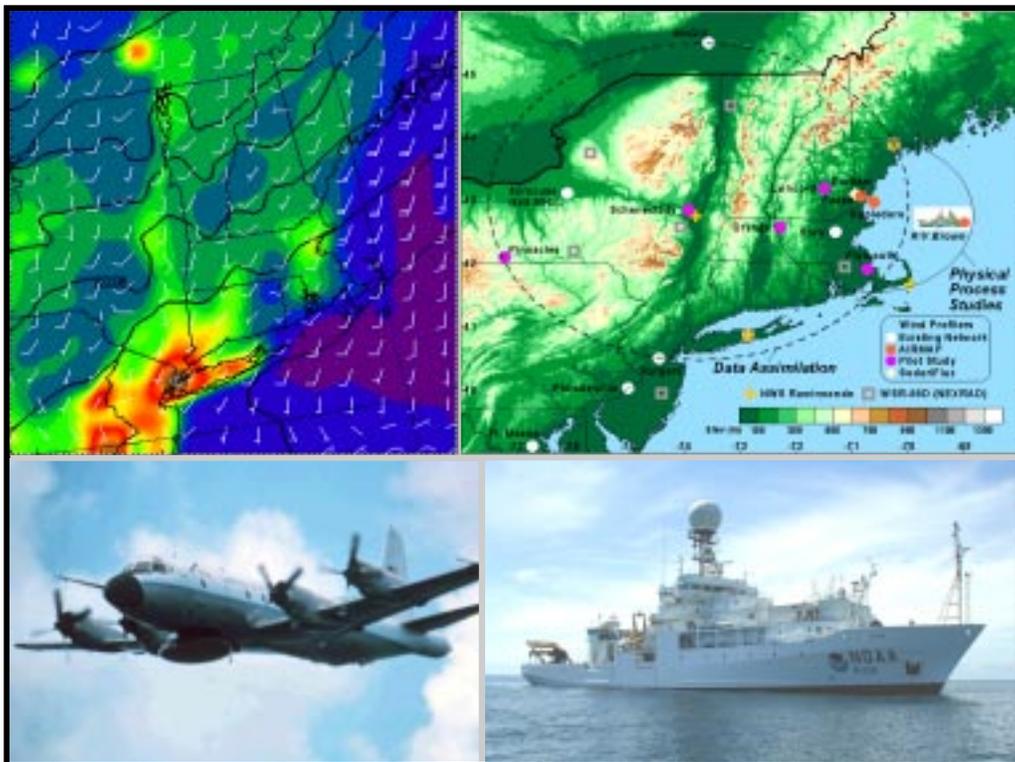




NEAQS-ITCT 2004

Science and Implementation Plan



NOAA's Atmospheric Research Campaign
NOAA's ARC
Combining Climate Change and Air Quality Research

April 2003

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Introduction

Scientific Motivation

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

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

During each of these intensives NOAA operated instrumented aircraft and participated in specialized ground based and remote sensing measurements. During the summer of 2002 NOAA deployed one of its research vessels, *Ronald H. Brown*, to the Gulf of Maine as part of the New England Air Quality Study (NEAQS), equipped with a comprehensive set of chemical and meteorological instruments to develop the information needed to plan the current effort.

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 either the starting point or the end point of regional air quality concerns depending on whether you are on the west coast (inflow) or east coast (outflow) of the U. S. Thus, in recognition of this strong linkage NOAA will conduct a joint regional air quality and climate change study in the summer of 2004. The study will combine the elements of the

previous ITCT and NEAQS studies and will be known as the **NEAQS-ITCT 2004**. The study will focus on air quality along the Eastern Seaboard and transport of North American emissions into the North Atlantic. The major NOAA assets (the two aircraft and the ship) will be deployed in a manner that will support the objectives of both research programs.

Previous Research

There have been at least three decades of studies that have been aimed, at least in part, at determining the causes of poor air quality outside of urban areas along the east coast of the U.S. and the transport of polluted air out into the North Atlantic. Several of the earliest studies [Zeller *et al.*, 1977]; [Kelleher and Feder, 1978]; [Spicer, 1982] presented 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 at Kejimikujik National Park in Canada [Brice *et al.*, 1988]; [Beattie and Whepdale, 1989], begun in 1979, demonstrated the transport of such plumes to central Nova Scotia, a distance of 500 km or more.

The Global Change Expedition/Coordinated Air-Sea Experiment/Western Atlantic Ocean Experiment (**GCE/CASE/WATOX**) [Pszenny *et al.*, 1990], carried out during the summer of 1988, was designed to study atmospheric and oceanic processes affecting the biogeochemical cycles of carbon, nitrogen, sulfur, and trace metals in the North Atlantic Ocean region. The stated objectives of the study included the identification of the factors controlling the ozone distributions in the North Atlantic marine boundary layer and the estimation of the contributions of sources on the surrounding continents to the sulfur, nitrogen and trace metal budgets of the region. Transport and deposition of aerosols is the dominant mechanism of continental contribution to these budgets.

A great many research vessel cruises through the Atlantic Ocean have included atmospheric measurements. [Winkler, 1988] has summarized the ozone measurements from 32 of these cruises conducted in 1977 through 1986. These data indicate that the surface ozone concentration is about twice as high in the Northern Hemisphere as compared to the Southern hemisphere, and this difference may, in part, be caused by transport of ozone photochemically produced. More recently, a comprehensive investigation of the O₃ and aerosol distributions over the North Atlantic was made during a cruise of the German research vessel Polarstern in 1987 [Special section, J. Geophys. Res, 95, D12, 1990].

Several NASA sponsored programs have identified transport of pollution from North America to the western North Atlantic. Airborne differential absorption lidar (DIAL) measurements [Harriss *et al.*, 1984] revealed persistent, highly stratified layers of aerosols extending more than 600 km into the Atlantic in the region between the U.S. east coast and Bermuda in July 1981 and August 1982. In situ measurements showed that these layers correlated with elevated ozone and CO levels. DIAL and in situ measurements of ozone, CO and aerosols [Wofsy *et al.*, 1992]; [Anderson *et al.*, 1993] off

the eastern coast of the U.S. have identified strong correlations between these species, and concluded that anthropogenic pollution has a major impact on the budgets of these species in the near continent region. [Fishman *et al.*, 1990];[Fishman *et al.*, 1991] have used satellite data to derive the tropospheric ozone distribution; a striking feature of their results is a strong, summertime maximum extending downwind from North America into the North Atlantic. This maximum is suggestive of transport of ozone produced photochemically in the troposphere.

Studies conducted in this region whose findings may be particularly important for planning and interpreting the 2004 study include the following. These can be roughly divided into rural northeastern North American continental studies and North Atlantic oceanic studies.

Rural northeastern North American continental studies

NACNEMS – During the late summer and early fall of 1988, measurements of many trace species of tropospheric photochemical interest were made at seven surface stations in the eastern U.S. and Canada that constituted the North American Cooperative Network of Enhanced Measurement Sites (NACEMS). The interpretation of the results was focused on the production of ozone and the partitioning of reactive oxidized nitrogen species in rural regions of eastern North America [Trainer *and al.*, 1993]; [Parrish *et al.*, 1993a]; [Roberts *et al.*, 1995]. Measurements continued after 1998 at some of the sites. In particular, the Whiteface Mountain site is still operating at present, and that site will participate in the 2004 field study. [Dutkiewicz *et al.*, 2000] report trends of SO₂ and sulfate over decades at this site.

The airborne Acid Model Operational Diagnostic Evaluation Study (ACID MODES) was loosely coordinated with NACEMS. This study was designed to provide data for validation of the regional acid deposition model (RADM). This study focused on processes in the gaseous phase and thus the flights were made mostly on sunny days with either clear skies or scattered cumuli. [Tremmel *et al.*, 1993];[Tremmel *et al.*, 1994] report measurements of peroxides from these flights.

Harvard Forest - Harvard Forest comprises approximately 3000 acres of land in Petersham, Massachusetts that include mixed hardwood and conifer forests. At the site, measurements of Net Ecosystem Exchange (NEE) for carbon, ozone, NO_y, H₂O, and energy have been made continuously for 10 years, along with comprehensive climatic, environmental, chemical, and ecological observations. Harvard Forest is ideally situated for a study of the pollution history of the Northeast urban/industrial corridor. As a trajectory analysis indicates, winds are predominantly from the west, with relatively clean, background air from the northwest (Canada) and more polluted air from the southwest (New York City—Washington, D. C. corridor). Winds from the east are rare (~10% of the time), typically occurring during stormy weather, and carrying an oceanic signal. Results from measurements at Harvard Forest that are particularly relevant to the 2004 study are reported in [Chin *et al.*, 1994]; [Goldstein *et al.*, 1995];[Goldstein *et al.*,

1996]; [Goldstein *et al.*, 1998; Hirsch *et al.*, 1996]; [Munger *et al.*, 1996]; [Munger *et al.*, 1998] [Liang *et al.*, 1998; Moody *et al.*, 1998] [Lefer *et al.*, 1999].

NARSTO-NE and NARSTO-NE-OPS – The program formerly known as the North American Research Strategy for Tropospheric Ozone (NARSTO) has conducted two field studies in northeastern North America. Little information regarding these studies can be found on the NARSTO web site. (<http://www.cgenv.com/Narsto/>). Some useful results from NARSTO-NE are reported by [Zhang *et al.*, 1998] and [Seaman and Michelson, 2000].

AIRMAP AIRMAP is a NOAA Cooperative Institute, a joint collaboration involving researchers in New Hampshire (University of New Hampshire, Plymouth State College, Mount Washington Observatory and the New Hampshire Department of Environmental Services) and NOAA's Aeronomy Laboratory (AL) and Forecast Systems Laboratory (FSL). AIRMAP is focused on atmospheric chemical and physical observations in rural and semi-remote areas of New Hampshire with the goal of understanding the inter-relationships in regional air quality, meteorology, and climate phenomena.

A network of rural air quality monitoring stations has been established under AIRMAP (Figure 1). These stations provide continuous measurements of ozone and fine particles and their precursor compounds as well as meteorological parameters in rural New Hampshire. Through the analysis of these data, a clearer picture of the factors

controlling air quality in New England is emerging [e.g., Slater *et al.*, 2002, Huiting and Talbot, 2003].

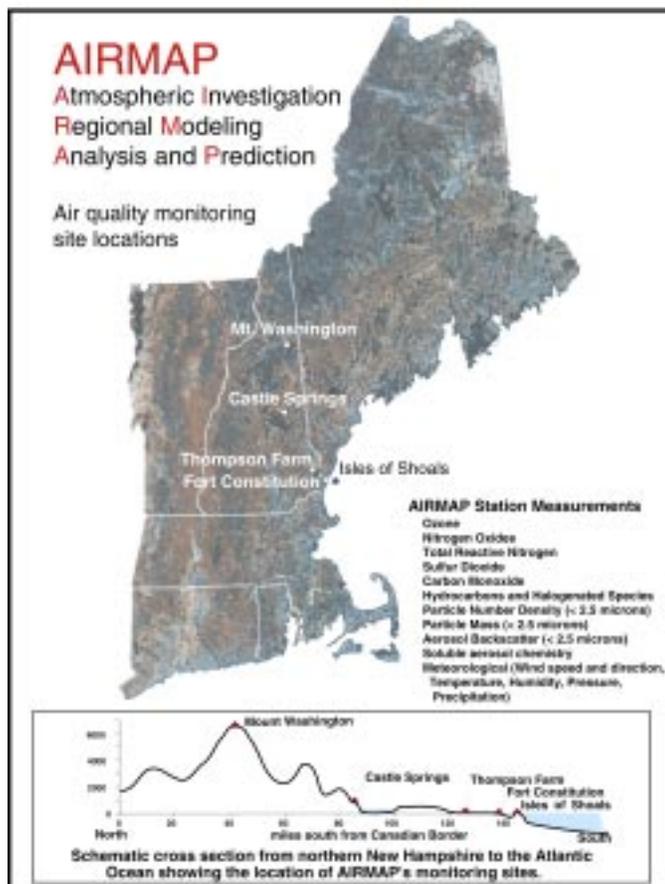


Figure 1. Map showing location of the AIRMAP air quality research monitoring network.

North Atlantic oceanic studies

AEROCE - A systematic study of the influence of anthropogenic emissions on ozone and aerosols over the North Atlantic was undertaken as part of the Atmospheric-Ocean Chemistry Experiment (AEROCE). This program began in 1988 and continued through 1998. Research focused on goals organized around two themes:

Theme 1: Ozone and oxidants - To understand the role of anthropogenic emissions and natural processes in the ozone budget and the oxidizing capacity of the troposphere over the North Atlantic Ocean.

Theme 2: Aerosols and climate - To characterize the chemical and physical properties of aerosols important to the radiative properties of the atmosphere and climate; to study the processes that affect these properties; and to assess the relative importance of natural vs. human sources.

The research was focused to a large extent around a series of long-term measurements carried out on the North Atlantic island stations of Barbados, West Indies; Bermuda; Tenerife, Canary Islands; Mace Head, Ireland; Heimaey, Iceland and Miami, Florida. These sites have continued to operate independently since AEROCE ended. A summary of the major accomplishments of AEROCE has been given by [Prospero, 2001].

ASTEX/MAGE - The Atlantic Stratocumulus Transition Experiment (ASTEX), one of the second series of FIRE international cloud-climatology experiments, took place in June of 1992 in the stratocumulus-capped marine boundary layer. The primary purpose of ASTEX was to study the factors influencing the formation and dissipation of marine clouds. IGAC's Marine Aerosol and Gas Exchange (MAGE) Activity organized the chemical experiment within ASTEX. Its objective was to study air-sea exchange and the formation and transformation of marine aerosols, in part by making Lagrangian observations (moving the measuring systems to stay with the same air). Chemical instrumentation was deployed on two islands, one French and two U.S. ships, and three aircraft from the U.S. and U.K. Although the meteorological situation was complex, chemists benefited from the observations of dozens of other groups who were studying cloud physics, boundary layer dynamics, and radiative transfer. They characterized turbulent and large-scale air motions in and above the boundary layer, thus enabling MAGE scientists to quantify the impact that dynamics had on chemical concentrations. A collection of MAGE papers has been published in a special issue of JGR: Volume 101, number D2, February 1996. Most of the more dynamically oriented ASTEX papers, many containing analyses based on the Lagrangian observational strategy, are contained in the August 15, 1995 issue of the Journal of the Atmospheric Sciences.

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

- To assess the long range transport of photochemically active compounds and/or their products and determine the impact of this transport on hemispheric air quality.
- To ascertain the effect of these compounds on the oxidative properties and radiation balance of the atmosphere.
- To estimate the amounts of these compounds that are deposited in this marine environment in order to better quantify the potential impact of this deposition on surface sea-water chemistry and marine biological processes.

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

Two additional field intensives were conducted in early spring, 1996 and late summer/early fall, 1997, which were reviewed in the IGACtivities Newsletter, Issue No. 24. The interpretation of these studies has focused on the chemical evolution, removal and transport patterns of anthropogenic emissions over the North Atlantic [Cooper *et al.*, 2001] [Cooper *et al.*, 2002b] [Cooper *et al.*, 2002a] [Stohl *et al.*, 2002], [Li *et al.*, 2002].

TARFOX - The overall goal of the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) is to reduce uncertainties in the effects of aerosols on climate by determining the direct radiative impacts, as well as the chemical, physical, and optical properties, of the aerosols carried over the western Atlantic Ocean from the United States. Subsidiary objectives of TARFOX are to:

- Perform a variety of closure studies by using over-determined data sets to test the mutual consistency of measurements and calculations of a wide range of aerosol properties and effects.
- Use the results of the closure studies to assess and reduce uncertainties in estimates of aerosol radiative forcing, as well as to guide future field programs on this subject (e.g., ACE-2).

TARFOX was conducted July 10-31, 1996. It included coordinated measurements from four satellites (GOES-8, NOAA-14, ERS-2, LANDSAT), four aircraft (ER-2, C-130, C-131, and a modified Cessna), land sites, and ships. A variety of aerosol conditions was sampled, ranging from relatively clean behind frontal passages to moderately polluted with aerosol optical depths exceeding 0.5 at mid-visible wavelengths. The latter conditions included separate incidents of enhancements caused primarily by anthropogenic sources and another incident of enhancement apparently influenced by recent fog processing. Spatial gradients of aerosol optical thickness were sampled to aid in isolating aerosol effects from other radiative effects and to more tightly constrain closure tests, including those of satellite retrievals. The results of TARFOX have been

published in a two special issues of JGR: Volume 104, number D2, January 27, 1999; and Volume 105, number D8, April 27, 1999.

ACE-2 - The anthropogenically influenced marine atmosphere of the North Atlantic was chosen for the second Aerosol Characterization Experiment (ACE-2). Aerosol characteristics, processes and effects were contrasted with those observed during ACE-1, which took place in the minimally polluted Southern Ocean. Four major aerosol types have been identified in the North Atlantic region: 1) sea-salt particles, ubiquitous in the lowest layers of the marine atmosphere, 2) dimethyl sulfide (DMS)-derived particulate mass, which has been identified throughout the marine troposphere, 3) Saharan mineral dust, predominantly found in the sub-tropical and tropical part of this area, and 4) anthropogenic aerosols, which have been observed over large areas of the North Atlantic Ocean. The goal of ACE-2 was to determine and understand the properties and controlling factors of the aerosol in the anthropogenically modified atmosphere of the North Atlantic and assess their relevance for radiative forcing. To achieve this goal, ACE-2 pursued three specific objectives:

Objective 1. Determine the physical, chemical, radiative and cloud nucleating properties of the major aerosol types in the North Atlantic region and investigate the relationships between these properties.

Objective 2. Quantify the physical and chemical processes controlling the evolution of the major aerosol types and in particular of their physical, chemical, radiative and cloud nucleating properties.

Objective 3. Develop procedures to extrapolate aerosol properties and processes from local to regional and global scales, and assess the regional direct and indirect radiative forcing by aerosols in the North Atlantic region.

The study was carried out from June 16 to July 25, 1997, and included 60 coordinated aircraft missions with 6 aircraft (for a total of 450 flight hours), one ship, and ground stations on Tenerife, Portugal and Madeira. The results of ACE-2 have been given in a special issue of *Tellus B*, Volume 52, Issue 2, April, 2000.

AEROSOLS99 - The Aerosols99 cruise crossed the Atlantic Ocean from Norfolk, Virginia, to Cape Town, South Africa, between January 14 and February 8, 1999. The goals of the cruise were to determine the chemical, physical, and optical properties of the marine boundary layer (MBL) aerosol, the vertical distribution of aerosols and ozone, the column-integrated aerosol optical depth, and the ozone, CO, and peroxy radical chemistry in the MBL. Sampling strategies were optimized to obtain data sets to evaluate satellite-derived ocean color (Sea-viewing Wide Field-of-view Sensor), aerosol optical depth (advanced very high resolution radiometer) and total column ozone (Total Ozone Mapping Spectrometer). The results of this study have been given in a special section of JGR, Volume 106, Issue D18, September 27, 2001.

PICO-NARE - A surface site in the central North Atlantic was established on Pico Island of the Azores in the spring, 2001 [*Honrath and Fialho*, 2001]. At an elevation of 2250 m, the site frequently samples free troposphere air. Year-round measurements of CO, NO_x, NO_y and black carbon are ongoing. Support to begin VOC measurements in the

spring of 2004 has been secured. The site is on the summit of Pico Mountain, an inactive volcano. Although the summit is an ideal sampling location, it presents logistical difficulties. The nearest road extends only halfway up the mountain, and only a steep footpath reaches the summit. Access is not possible during winter months. The site was set up with helicopter support, and power is provided via a 3 km medium-high-voltage cable to a small generator 1000 m lower in altitude. All instruments are automated, and the site is controlled and monitored over the internet using the cellular telephone network.

The New England Air Quality Study (NEAQS) 2002 – During the summer of 2002 NOAA deployed one of its research vessels, *Ronald H. Brown*, to the Gulf of Maine. The ship was instrumented with a full array of instrumentation to quantify gaseous and aerosol pollution and important meteorological parameters. The ship was used to study the chemical evolution of the New York city and Boston plumes and to track pollution being transport into the Gulf of Maine and into coastal New England. These measurements were coordinated with intensive measurements in the AIRMAP research network and airborne measurements conducted with the DOE G-1 research aircraft. The study intensive was conducted from July 12 through August 10.

Research Areas

The research planned for the 2004 field campaign has been organized around the following five research areas, each with an associated science question.

Emissions verification - How well do current inventories represent actual emissions for: cities, point sources, ships, and vegetation?

Transport and mixing – What are the relative amounts of pollution imported to New England and exported from the continental boundary layer to the marine boundary layer and the free troposphere

Chemical transformation – How do gaseous and aerosol emissions evolve chemically and physically as they are transported away from the source regions to the remote atmosphere?

Aerosol properties and radiative effects – What are the chemical, physical, and optical properties of the regional aerosol and how do these properties affect regional haze and aerosol direct and indirect radiative forcing of climate?

Forecast models – What is the current skill of air quality forecast models on local, regional and global scales and what improvements can be made to enhance the accuracy and extend the periods of these forecasts?

The specific goals and planned approach for each of these research areas is described in more detail in the following sections.

Emission Verification

Relevance

Improving the quantitative understanding of the location, timing, and speciation of chemical and aerosol emissions into the atmosphere is critical to advancing the knowledge of tropospheric chemistry, transport, and transformation on a variety of spatial scales. Known or suspected inaccuracies in current emissions inventories of both anthropogenic and biogenic sources account for a substantial amount of the total uncertainty in model simulations of air quality and climate change processes [IPCC, 2001]. Reports suggest that while some U.S. inventories are reasonably accurate (e.g., point source NO_x and SO₂: [Ryerson *et al.*, 1998]), others may have substantial errors (e.g., urban CO: [Parrish *et al.*, 2002]; petrochemical alkenes: [Wert *et al.*, 2003]). A goal of the NOAA 2004 summer field intensive is to use ambient measurements to better constrain the emissions inventories of anthropogenic and biogenic compounds relevant to both regional air quality and climate change.

Science questions

Quantitative information on the emissions from a variety of point and area sources is required to understand their relative impacts on the atmosphere, both in terms of air quality and of radiative effects from aerosol formation. Substantial contributions from mobile sources (primarily automobile and truck traffic) and from large electric utility power plants to anthropogenic gas- and aerosol-phase pollutants in the summertime New England airshed are expected. Other potentially significant contributions may come from emissions from industrial processes and/or commercial shipping. Substantial biogenic contributions to reactive VOC compounds involved in ozone and secondary organic aerosol formation are also expected.

The summer 2004 intensive will provide data that will help answer the following basic questions for a variety of source types. Direct emissions of a variety of species will be studied, including aerosol (e.g., black carbon (BC)) as well as gas-phase (e.g., VOCs, SO₂, CO, etc.) compounds.

1. *How well are the ratios of co-emitted pollutants represented in existing emissions inventories?* Many sources emit more than one compound at a time, often in characteristic ratios that can be used to identify a particular source among several possible emitters. Sources with differing emissions ratios, such as for (VOC/NO_x), can have substantially different impacts on the rates of photochemical transformations occurring downwind. Spatial patterns of transport and deposition of secondary photoproducts and aerosol particles may also depend on ratios of direct emissions from a given source. Thus, more accurate estimates

of emissions *ratios* will improve the ability to predict the various impacts of a given source on the atmosphere downwind.

2. *How well are the absolute amounts of emitted pollutants represented in inventories?* Model-measurement differences can arise due to inaccuracies in the source emissions rates used as model input. Actual emissions rates can vary substantially on timescales of hours, days, seasons, or years, some of which may not be captured by a given inventory compilation. Improved estimates of the absolute amounts of emissions from a variety of different source types will minimize this potential source of model-measurement bias, and enhance the utility of models to usefully explore future “If ... then...” emissions control scenarios.
3. *How well are the spatial patterns of emissions represented in inventories?* As an example, the geographic distribution of SO₂, NH₃, and biogenic terpenoid emissions will determine the relative contribution of anthropogenic sulfate to biogenic secondary organic aerosol for a given receptor region. The location and magnitude of aerosol black carbon (BC) sources, primarily from urban areas, will influence the radiative impacts of transported emissions downwind. Regional surveys will provide data to evaluate and improve the spatial accuracy of current inventories for both air quality and climate-relevant species.
4. *How well are temporal variations in emissions strength and composition represented in inventories?* Biogenic emissions of isoprene and monoterpenes are highly dependent on sunlight, temperature, and drought conditions. Anthropogenic emissions can also vary substantially across a range of timescales. Variability in either can influence the chemical composition and radiative properties that result from such emissions into the atmosphere. *In-situ* measurements over the course of the 2004 summer field intensive will provide information on emissions variability on timescales of hours to weeks, covering the range of ambient conditions occurring during the study period.

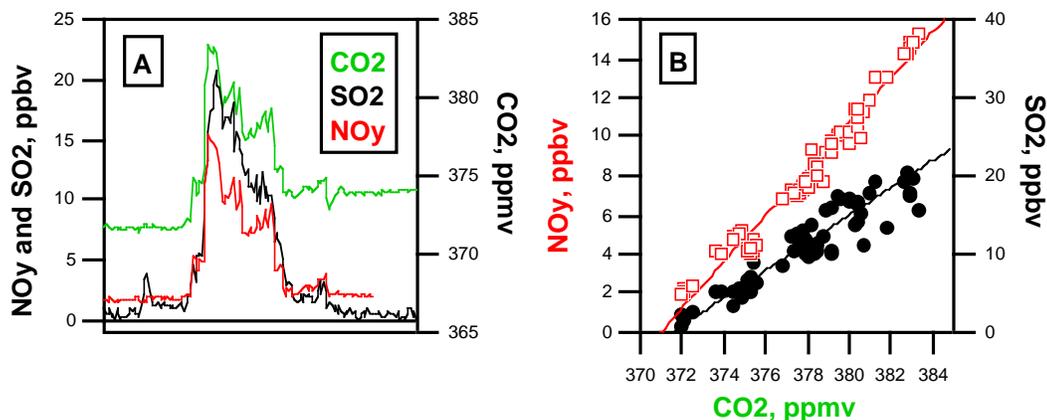
Deployment strategy

Comparison of ambient data to emissions inventories has been an integral part of NOAA tropospheric field programs in the past. The three NOAA-sponsored mobile platforms – the WP-3D, the ETL lidar aircraft, and the R/V *Ronald H. Brown* – will all acquire data suitable for emissions inventory comparison in the course of pursuing other scientific objectives. As such, this research area is relatively easily integrated within the overall requirements of regional air quality and climate change research, and will nicely complement the other deployment activities described in this document. Some examples of ship or aircraft tracks that will generate data suitable for emissions inventory assessment are briefly described below.

Emissions ratios

Data generated in near-field plume transects within or immediately downwind of source regions can be used to assess emissions *ratios* of pairs of co-emitted compounds. Near-field transects reduce the uncertainty in deriving emissions *ratios* by minimizing the effects of differential removal rates and of plume dilution into different background abundances of the two species in question. Near-field transects also enhance the observed mixing ratio enhancements above background, maximizing the signal-to-noise for the two species; given substantial in-plume enhancements, to first order, dilution will be equal for both co-emitted species. The slope of a two-sided linear regression fit to the plume transect data will give a direct measure of the emissions ratio. Under these conditions, the uncertainty of the emissions ratio derived from measured data is determined by the combined uncertainty of the individual instrument calibrations. For well-operated instruments, this uncertainty can be less than $\pm 10\%$, providing an independent and accurate check on the ratio calculated from an emissions inventory.

Appropriate data can be generated by a single WP-3D traverse of a plume within the boundary layer; typically, this is repeated to build statistics, to capture temporal



	<u>Obs.</u>	<u>CEMS</u>
NO _x /CO ₂	1.2±0.1	1.4±0.1
SO ₂ /CO ₂	1.8±0.2	1.9±0.2

Figure 2. (A) An 18-km horizontal segment of NO_y, SO₂, and CO₂ data downwind of the Limestone power plant in east Texas on July 10, 2000. (B) Two-sided regression fits to data from (A). The table compares the derived emissions ratios to reported values using data from the continuous emission monitoring system (CEMS) installed at the plant.

variability, and to ensure that differential lofting of emissions (e.g., from multiple sources at different release heights) can be taken into consideration. Alternately, these data can be generated by locating *Ronald H. Brown* downwind of an emissions source and sampling normally. Atmospheric variability will cause directly emitted species to co-

vary over time, generating a trend line with a characteristic slope reflecting the ratio of two species at the time of emission. The data from the ship and from the aircraft should result in derived emissions *ratios* that are identical, within the stated uncertainties, for a given plume. As long as the assumption of negligible differential loss is met, emissions *ratios* estimated from ambient measurements provide a robust and independent benchmark value against which emissions inventories can be validated.

Using the above analysis, the geographic and temporal distributions of emissions can be evaluated from measurements and compared to available inventories. Highly time-resolved data from continuous emissions monitoring systems (CEMS) can be directly compared to ambient plume measurements taken concurrently. Comparison of accurate but annually-averaged inventory values to ambient data taken on shorter time scales can provide information on the expected range about the averaged tabulated value.

Absolute emission rates

Knowledge of the magnitude of emissions from a single source is more difficult to extract from ambient measurements. While emissions *ratios* are relatively insensitive to dilution, deriving *absolute emission rates* from ambient data requires that dilution be taken into account in the calculation. One way to derive emission rates uses a mass balance approach, in which the total number of molecules in an emissions plume is calculated and compared to the reported emissions rate (e.g., [White *et al.*, 1976]). Additional assumptions inherent in this analysis increase the resulting uncertainty above that for calculating emissions ratios. Given the large potential errors, these assumptions must be evaluated quantitatively for a given plume. Under ideal meteorological conditions, however, the uncertainty in deriving absolute emissions rates from ambient measurements appears to be $\pm 25\%$ or less, which is sufficiently low to provide a useful check on point source emissions inventories.

Data used to calculate absolute emission rates are taken in a manner similar to that employed for calculating emission ratios. Near-field transects of large point or area sources, repeated at different altitudes and distances downwind, will be carried out to generate sufficient data to calculate absolute emission rates as well as to quantify the assumptions inherent in the analysis. If logistics permit a joint aircraft mission, the ETL profiling lidar aircraft could provide important information on plume vertical structure and boundary layer height while the WP-3D performs plume transects within the boundary layer.

Transport and Mixing

Relevance

Air quality in the continental and marine regions of New England is highly dependent on the meteorological conditions that govern the transport and mixing of trace gases and aerosol particles. These processes occur on a variety of scales: atmospheric boundary layer (ABL), regional, intracontinental and intercontinental. Previous studies have demonstrated, on all of these scales, the impact of transport processes on east coast air quality. NARE has shown that major east coast pollution episodes develop on a regional scale under stagnant surface high pressure conditions and are subsequently exported to the western North Atlantic Ocean in the warm sector of mid-latitude cyclones [Merrill and Moody, 1996]. Figure 3 shows the median ozone mixing ratios exported from North America during late summer within the various airstreams of mid-latitude cyclones. Export to the North Atlantic lower troposphere is influenced by boundary layer processes, and the pollution can either be confined to near-surface flow in the marine boundary layer as was shown during NEAQS 2002 [W. Angevine, personal communication], or transported aloft in layers that become liberated from the stable and shallow marine boundary layer [Angevine *et al.*, 1996]. On the broader, intracontinental scale, Wotowa and Trainer [2000] have shown that large quantities of carbon monoxide from Canadian forest fires can impact the eastern United States during summer. And on the intercontinental scale pollutant transport from Asia and Europe is believed to have substantial impact on background ozone mixing ratios across the United States [Fiore *et al.*, 2002].

In contrast to earlier studies conducted along the east coast and New England, the 2004 study is being implemented to examine the full range of transport scales and their influence on New England air quality. Attention will also be given to the intra- and intercontinental export of pollution from the New England region, focusing on transport to the Canadian Maritimes the North Atlantic Ocean and Europe. A clear understanding of transport on these scales will allow us to assess the impact from local, regional and distant sources on the air quality of air masses as they pass through the New England Region and are exported to downwind locations.

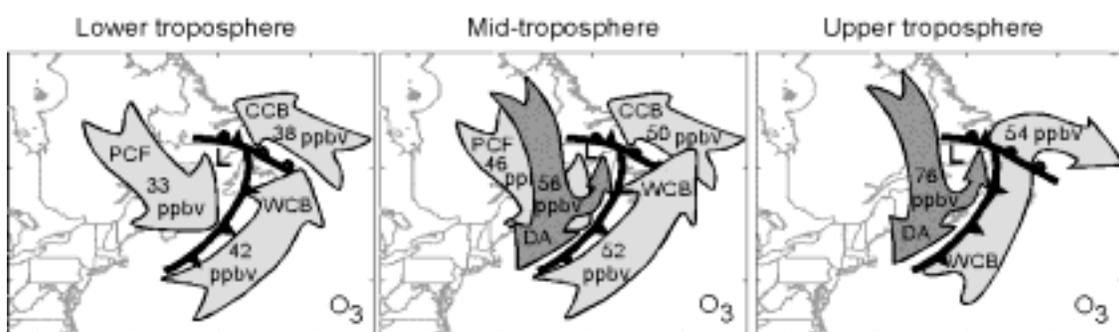


Figure 3. Median ozone mixing ratios (ppbv) associated with the various airstreams of mid-latitude cyclones: warm conveyor belt (WCB), cold conveyor belt (CCB), post cold front airstream (PCF), and the dry airstream (DA). Measurements made during late summer, 1997, by the NOAA WP-3D aircraft during the NARE'97 experiment [Cooper *et al.*, 2002].

Science Questions

Atmospheric boundary layer scale

Much of our understanding of atmospheric boundary layer scale mixing and transport processes in the coastal region of New England has emerged from the NARE 1993 campaign. *Angevine et al.* [1996ab] describe measurements made at the Chebogue Point coastal site in Nova Scotia using a wind profiler and chemical measurements made at the surface and from aircraft. Similarly, *Knapp et al.* [1998] describe ozone and meteorological measurements made from a parafoil kite at Cape Sable Island, Nova Scotia. These studies detected stratified pollution layers exported from the continent to the marine atmosphere and identified the transport processes that formed these layers. The NEAQS 2002 study has explored these processes further but the results are not yet available. Findings from these studies have led to the formulation of three science questions that re-visit the basic ABL transport and mixing processes already identified but with the goal of refining our understanding of their spatial and temporal scales, especially in the transition zone between the continental and marine regions.

1. *What processes control the mixing and layering of pollutants within the ABL?* Pollutants are often well mixed throughout the ABL during daytime over land but are typically transported as layers at night and above water. Measurements from wind-profilers, rawinsondes, *in situ* measurement surface sites, the NOAA aircraft, and lidar and flux sensors on *Ronald H. Brown* will provide information on the depth, layering, turbulence characteristics, and chemical composition of the ABL. These data, gathered over a wide area, will allow researchers to determine the timing and location of pollutant transport via mixing or layering. Researchers will also be able to identify the conditions that either isolate polluted air from the surface or bring it directly into contact with humans and vegetation at the surface.
2. *What processes control pollutant transport at the land-sea interface?* Coastal topography and land/sea breezes influence ABL pollutant transport in coastal regions. Well-mixed pollution in the ABL over land can become layered as it is transported over water, conversely layered pollution over water can mix throughout the ABL when it comes ashore. Using the same measurement platforms mentioned above researchers will assess the impact of coastal processes on the pollutant distribution within air masses that cross the land-sea interface, and will investigate the associated time scales.
3. *Where and when is polluted air imported to and exported from the ABL?* Pollution from upwind sources that impacts the surface of the New England region must at some point become entrained into the ABL above New England. This imported pollution plus that emitted from within New England will either be wet or dry deposited at the surface or will eventually be exported from New England. The mass fluxes in and out of the New England ABL will either occur laterally at low altitudes, or through vertical motions and mixing with the free

troposphere above New England. Researchers will investigate the location and timescales of the convective processes that mix air in and out of the ABL. Particular attention will be paid to convective processes at times when pollution within relatively cold air advects over the warmer waters of the Gulf Stream. Another key process to explore is the lofting of pollutants as they pass over the mountainous regions of New England.

Regional and Intracontinental Scale

The NARE campaigns during the 1990's have provided several years of surface chemical measurements in the western North Atlantic region in addition to aircraft campaigns conducted during all four seasons along the North American east coast and western North Atlantic. The results from these studies have provided a basic understanding of the regional and intracontinental scale transport processes that influence the region. For example, *Parrish et al.* [1998] describe surface ozone and CO measurements at several locations in the western North Atlantic linking the most polluted air to southwesterly transport from North America. *Cooper et al.* [2002] analyzed NARE aircraft flights and classified the measurements according to their location within mid-latitude cyclones to identify typical trace gas signatures of the various cyclone airstreams. The export of NO_y from North America and its subsequent removal is discussed by *Stohl et al.* [2002]. And outside of the NARE framework, *Moody et al.* [1998] identified the regional scale flow patterns associated with clean and polluted air measured at Harvard Forest, Massachusetts. These earlier works have identified the regional scale transport processes that impact the study region but estimates of the quantity of pollution impacting New England from various source regions are still lacking. The following science questions were developed with this short-coming in mind and are also focused on exploring undocumented regional transport processes.

4. *What source regions across North America impact New England air quality and what are the associated transport pathways and meteorological mechanisms?* Modeling studies using CO tracers, tagged according to the various emissions regions of North America, will be useful tools for answering this question. Particular attention will be paid to emissions from the Ohio Valley, SE USA, the Mid-Atlantic States, NE USA, the populated regions of Ontario and Quebec and Canadian forest fires. In terms of transport mechanisms, mid-latitude cyclone warm sectors will be studied but undocumented pathways will also be explored. The modeling studies will also focus on the quantity of pollution in the New England region that originates within the various upwind source regions.
5. *Can cold air outbreaks from Canada transport pollution from eastern North America to the Gulf Stream where convection could loft pollution from the lowermost troposphere to faster transport layers aloft?* Post-coldfront outflow would be the most likely mechanism for transporting Canadian forest fire emissions to New England and these air masses would also contain fresh emissions from eastern Canada and the northeastern USA. Cold frontal passages are common in this region during summer and regularly transport air to the

western North Atlantic. Whether or not the air remains cold enough to trigger convection and the lofting of pollutants over the warm waters of the Gulf Stream remains to be seen.

6. *How much of an impact do stratosphere/troposphere exchange events have on summertime ozone episodes across North America?* Stratospheric intrusions are most intense during winter and spring with a much lower intensity in late summer and early autumn. Nevertheless, stratosphere/troposphere exchange will still occur over North America during the study period. Exchanged stratospheric ozone can descend through the high pressure systems that produce the intense pollution episodes over the eastern USA and therefore can become entrained into the ABL.

Synoptic and Intercontinental Scale

While many studies referenced above and in the Introduction describe the export of pollution from the New England region to the western North Atlantic Ocean, to-date no study has specifically addressed the export of New England air masses to other continents. However, guidance on this issue can be taken from several recent papers by *Stohl. et al.* [1999, 2003ab] on pollutant transport from eastern North America to the atmosphere above Europe. In each case the intercontinental transport occurred within a warm conveyor belt. In terms of pollution from upwind continents being transported to New England, no cases have so far been documented. However, several recent papers have described the transport of Asian pollution to the west coast of North America [*Jaffe et al.*, 1999, 2003; *Cooper et al.*, 2003; *Forster et al.*, 2003]. For the proposed study, these Asian pollution events will have to travel an additional 3000 km to the New England study region and discerning between these air masses and polluted North American air masses may be difficult.

7. *Which source regions outside of North America have an impact on New England air quality and what are the associated transport pathways and meteorological mechanisms?* Results from ITCT 2002 have shown that plumes of Asian pollution can be transported rapidly to North America, but it is unknown if similar transport occurs in summer and if the Asian pollution can significantly impact the surface of the New England region. Warm conveyor belt (WCB) transport is probably the only mechanism that can transport a coherent plume from Asia to New England but WCB transport during summer is fairly weak. Modeling studies using tagged CO tracers from the different continents can identify instances of intercontinental pollutant transport and estimate the quantity of pollution that reaches the surface. The existence of these plumes will be verified with *in situ* aircraft measurements.
8. *What are the major pollution transport pathways from North America and where do these plumes go?* Pollution in the New England Region is eventually exported from North America, if it isn't wet or dry deposited first, along with pollution from other regions of North America. A specific goal of this study is to intercept

a plume of pollution as it forms in the ABL of North America, and then intercept the plume as it leaves the ABL and travels across the Atlantic towards Europe. WCBs are the most likely transport mechanism but others will be explored if they become evident.

Deployment Strategy

Answering the ABL scale Science Questions requires the deployment of surface measurement sites, the R/V *Ronald H. Brown* and NOAA aircraft. Over land the radar wind profiler network will play a key role (see the section on the profiler network) in determining the spatial and temporal scales of ABL processes, and ceilometers and chemical measurements at other static ground sites will also provide key information. To characterize the marine boundary layer *Ronald H. Brown* will be equipped with *in situ* aerosol and chemical measurement instruments, an ozone lidar, a Doppler lidar, a wind and temperature profiler, rawinsondes, and flux instruments.

In terms of the deployment of *Ronald H. Brown*, new ship patterns focusing on medium range transport are required, in addition to the successful patterns used in NEAQS 2002, which concentrated on short-range transport. The ability of the ship to "chase" pollutant plumes is limited by its low speed, so we must either take advantage of longer periods of near-stationary transport or predict where plumes can be found. Three basic patterns are proposed, one each to study the structure of pollution plumes as they intersect the downwind coast, as they traverse the Gulf of Maine, and as they traverse the warmer waters of the Gulf Stream.

- New England coast: Southwesterly or southerly flow would be required. The ship should cruise generally along the coast repeating a 20-40 km track. The specific region should be chosen based on predictions of maximum pollutant concentration, but would either be southeast of Boston where plumes leave the continent, or further downwind along the southern Maine coast where the plumes would re-enter the continental ABL.
- Gulf of Maine: To avoid sampling its own plume the ship would have to travel from the southern Maine coast southeastwards into the Gulf of Maine and the western North Atlantic under conditions of southwesterly flow. A ship track 150-200 km long would provide for characterization of boundary layer structure extending over the coldest waters. Surface-based pollutant layers may be found, but lofted layers are more likely.
- Gulf Stream: The ship would transit into the nearest portion of the Gulf Stream to look at how the boundary layer is modified by transport over warmer water. This pattern would be appropriate for a northwesterly flow period (Figure 4).

The NOAA WP-3D and NOAA ETL ozone lidar aircraft should be deployed in coordination with *Ronald H. Brown* to probe the depth of the marine boundary layer and the extent of any vertical mixing over the Gulf Stream. The lower troposphere can be explored on missions designed to chase ship plumes or to study the chemical

transformation of large plumes exported from North America. But a key role for the aircraft will be to explore the lateral as well as vertical extent of the plumes.

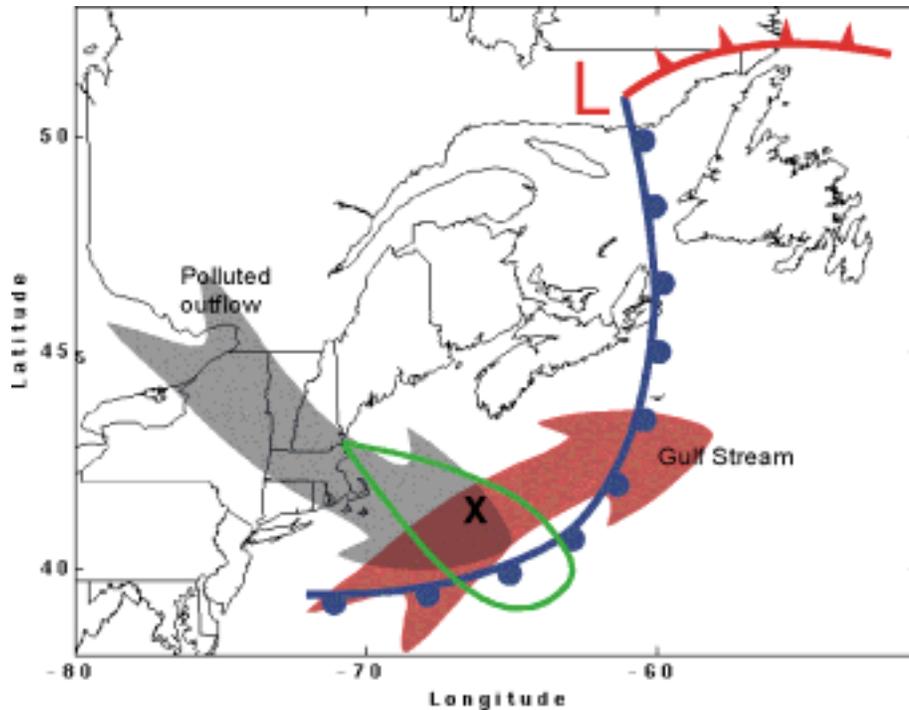


Figure 4. Flight plan schematic for a scenario in which fresh North American emissions (gray arrow) are advected over the warm waters of the Gulf Stream (red arrow) at low altitudes behind a cold front. Also shown are the locations of the aircraft flight track (green line) and *Ronald H. Brown* (black X).

To answer the regional and intracontinental scale Science Questions, the WP-3D would be flown through a variety of airstreams and across as many fronts as possible, between altitudes of 0 and 8 km above the continental and marine regions of New England. Upwind emissions source regions should also be explored. The NOAA ETL ozone lidar aircraft would follow a similar route, focusing on the lower troposphere. This approach will provide the greatest opportunity to characterize the various air masses (and their source regions) impacting the study region (Figure 5). Dispersion model forecasts can be used to steer the aircraft into air masses from various source regions. It will also be important to plan the flight tracks so that they capture the full width and vertical depth of the plumes.

It is anticipated that NOAA CMDL will launch daily ozonesondes from several locations across the United States. Possible sites are Trinidad Head, Boulder, Rhode Island and Sable Island. These measurements will yield important information on the ozone gradient across the United States. The data will be analyzed to determine ozone source regions and the contribution of transport patterns to the ozone gradient.

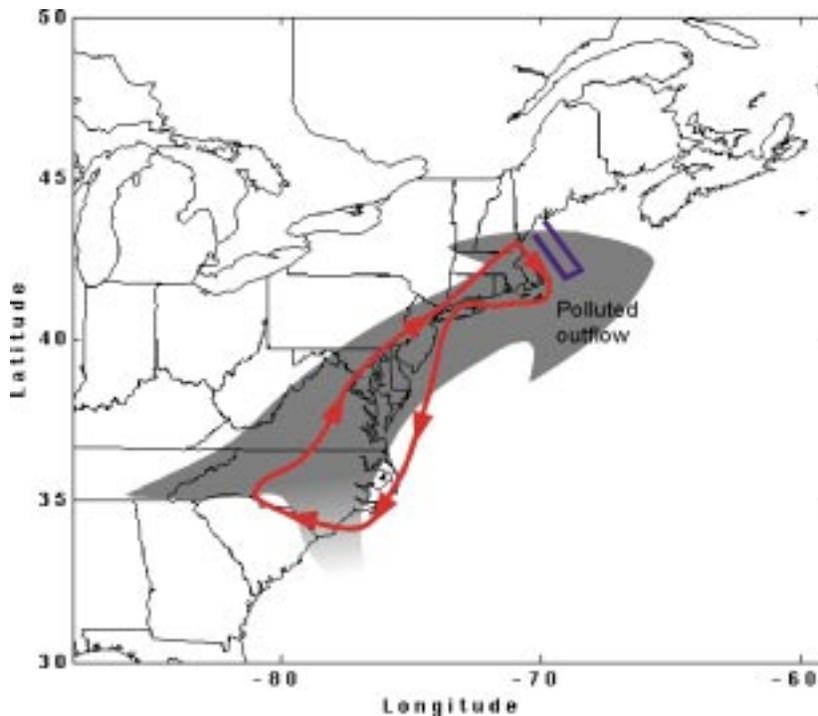


Figure 5. Flight plan schematic showing polluted continental outflow from the southeastern USA and New England. Also shown are the aircraft flight track (red line) and the ship track (purple line).

Finally, to answer the synoptic and intercontinental scale Science Questions the NOAA WP-3D and NOAA ETL aircraft will need to be steered into pollution plumes predicted by the dispersion models. This will be fairly straightforward for plumes from upwind sources but much more difficult for sampling a plume repeatedly from its formation in the North American ABL and during its subsequent transport across the Atlantic to Europe. Software routines will have to be developed such that a dispersion model can forecast a plume to travel from the North American ABL to Europe. It must then report the vertical and horizontal location of the plume at times when it can be sampled in the North American ABL, and at times when it can be intercepted by the aircraft based in the Azores and Scotland. Sampling the same portion of the plume will be crucial to understanding the dilution and chemical transformation processes that occur between North America and Europe (Figure 6).

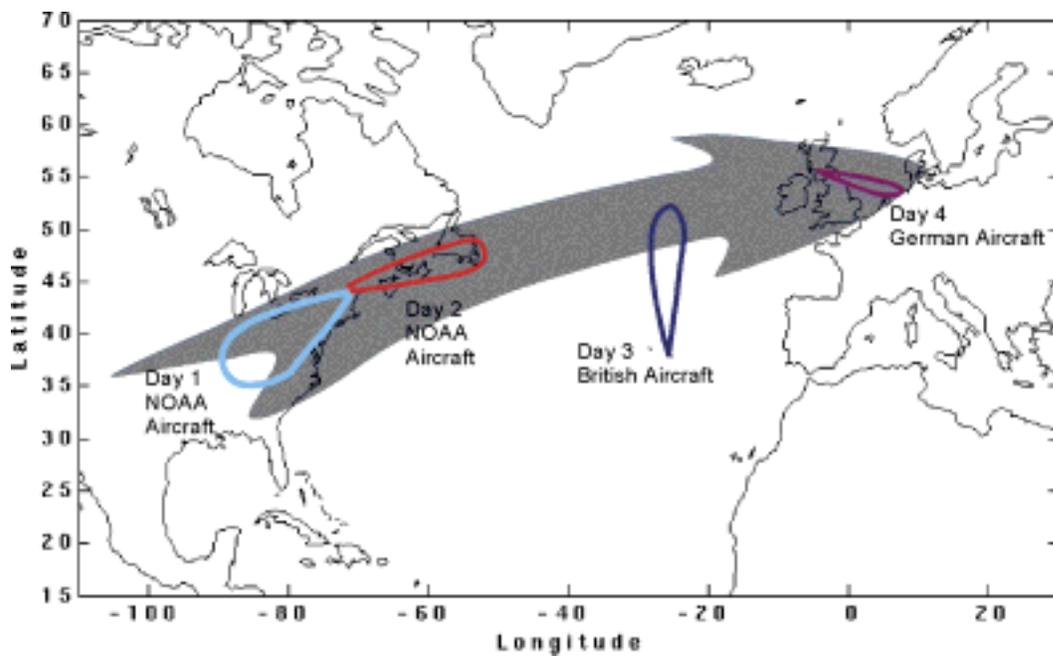


Figure 6. Schematic diagram showing the path of a polluted air mass advected from North America to Europe (gray arrow) and the proposed flight tracks for the NOAA, British and German aircraft.

Chemical Transformation

Relevance

Ozone and aerosols are harmful to humans and their presence in the atmosphere can influence regional and global climate. As a consequence, it is important to understand the factors that determine the distribution of ozone and aerosols in the atmosphere and that imbue aerosols with the chemical properties that impact climate and make aerosols deleterious to human health.

The section on “Emission Verification” discussed research that will be undertaken to better identify and quantify the sources responsible for the direct emission of aerosols and the sources of the chemicals responsible for the secondary production of ozone and aerosols in the study region. The section on “Transport and Mixing” indicated the research to be undertaken to understand the processes that mix ozone and aerosols from these sources through the atmosphere and that transport those compounds from the source regions and deliver them to receptor locations. This section describes the research that will be undertaken during the study to better understand the chemical processes that lead to the formation and loss of ozone in the atmosphere, the formation of aerosols and their transformation during transport.

The proposed research is based on the recognition that the principle factors that shape daytime chemistry that leads to ozone formation is reasonably well known but the chemical processes involving ozone during the nighttime and the importance of these processes is much less certain. By contrast the processes responsible for the formation of aerosols in the atmosphere and the transformation of the aerosols during transport is even less certain.

Science Questions

The summer 2004 intensive will provide data that will help to address two overarching questions relevant to regional air quality and global climate:

1. *How do emissions from local and distant source regions interact to determine the air quality in New England?*
2. *Can we follow the chemical evolution from the source regions to the source free North Atlantic?*

The mosaic of ozone and aerosol precursor emissions that influences the air quality in New England provides the opportunity to investigate the chemical evolution of individual source categories and their synergism in targeted flights. Three distinct distant source regions influence the air quality in New England:

- The eastern-seaboard, urban corridor that extends from Washington to Boston,

- Strong point source emissions along the Ohio River, and
- Industrial and urban emissions from the upper Midwest, including Chicago, Detroit and Toronto.

The chemical compositions of air masses that originate in these regions and reach the New England have been characterized.

Analysis (c.f., Moody et al., 1998) of measurements made at the Harvard Forest site in Central Massachusetts have identified the impact of various source regions on air quality in that region. During the daytime in the summer, the highest ozone and oxidant concentrations are generally observed under stagnant conditions when nearby local emissions accumulate, or when transport from the southwest permits pollution from the large urban sources along eastern seaboard to reach the site. Air-masses transported to these sites from the industrial/urban Midwest also have concentrations of ozone substantially greater than background and indicate a much greater degree of chemical processing. In contrast, air masses that are transported from the north and northwest generally bring clean air. However, the measurements at the AIRMAP sites showed that this could change dramatically in the event of forest fires in remote regions of Canada.

The aerosol compositions from the three source regions are expected to be distinct. Pollution from the eastern seaboard should be characterized by higher concentrations of organic aerosols due to primary emissions in the urban areas and the formation of secondary organic aerosols produced by the oxidation of anthropogenic volatile organic compounds, in particularly long chain aromatics. The aerosols contained in air transported to the region from the Midwest contain a much larger fraction of sulfates. The composition of aerosols transported from the more remote regions to the north and northwest may contain a higher fraction of natural VOC oxidation products. During NEAQS 2002 when winds were from the northwest, measurements made on the Ron Brown in the Gulf of Maine sailing off the coast of northern Maine suggested a connection between natural hydrocarbons and organic aerosols.

The export of photochemically produced ozone and aerosols to the North Atlantic has been seen at Seal Island and Chebogue Point at the southern tip of Nova Scotia and at Sable Island, as well as in the airborne measurements during the NARE experiments in 1993, 1996, and 1997. The measurements at the surface sites show that highly processed air reaches these sites that are about 500 km and 1000 km from the emission sources along the east coast. The measurements show that a large fraction of the reactive nitrogen species has been removed during the long-range transport within the marine boundary layer. Nevertheless, a model analysis by Duderstadt et al. (1998) of the measurements at Sable Island has shown that the NO_x concentrations during transport events are still sufficient to sustain net ozone production. These finding, if general, would suggest that active photochemistry during long-range transport can form significant ozone and secondary aerosols.

Based on present understanding, the study will focus on specific science questions.

Chemistry That Shapes Atmospheric Composition in New England

Daytime: It is now generally accepted that a reasonably well-defined, odd-hydrogen photochemistry controls the chemical processing during the daytime. It is more important to test how well this understanding is being translated into chemical/dynamical models that simulate daytime conditions. During the daytime over the continent plumes will be well mixed within the turbulent daytime boundary layer. As the plumes travel from the sources, they will interact with natural and anthropogenic emissions along their trajectory. This will provide opportunity to study the chemistry with various NO_x to VOC ratios at various stages of photochemical oxidation.

1. *What are the processes that control the rate and efficiency of ozone and secondary aerosols formation during the daytime downwind of power plants and urban plumes over regions with different natural hydrocarbon emissions?*
2. *During the daytime, how efficient are natural hydrocarbons like isoprene and monoterpenes in forming ozone and aerosols?*

Nighttime: The chemical processing that occurs during the nighttime hours is much less well understood. The ability of models to properly simulate the full diurnal cycle must be evaluated. The study will address the importance of the nighttime oxidation as a loss process for NO_x and investigate its influence on ozone production. Measurements over different source regions will provide important new information concerning loss mechanisms for NO_x at night. However, understanding the chemistry over the continent

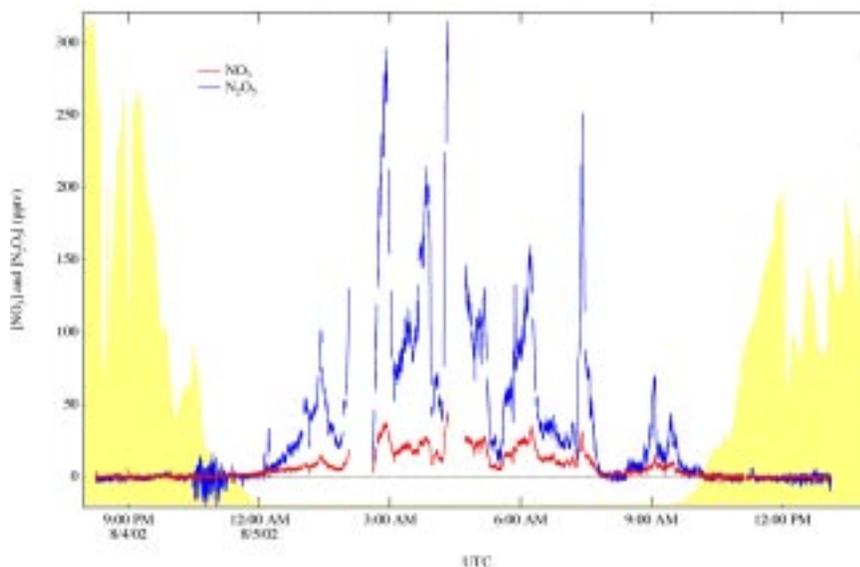


Figure 7. Concentrations of NO_3 (red trace) and N_2O_5 (blue trace), expressed as a mixing ratio in parts per trillion (pptv), measured aboard *Ronald H. Brown* during NEAQS 2002 with the *in-situ* cavity ring-down instrument. The data are for the night of August 4 - 5, 2002, and the x-axis is in universal time. The yellow background shows the intensity of shortwave radiation indicating the times of sunset and sunrise. Measured concentrations are below the instrumental detection limit (0.5 pptv) during daylight hours, but are nonzero and variable during the night, depending on the concentrations of other chemical constituents.

during the night will present a significant challenge due to the layering of the atmosphere.

3. *What are the processes that control the rate and efficiency of ozone and secondary aerosols formation during the nighttime downwind of power plants and urban plumes?*
4. *What role do biogenic VOCs play in nighttime chemistry? Are the reactions of monoterpenes with ozone and/or NO_3 radicals at night an important mechanism for aerosol formation.*

Chemistry Over the North Atlantic

Marine boundary layer: The chemistry that occurs in the polluted marine boundary layer is uncertain. The effect of this chemistry depends on the stability of the marine boundary layer and the residence time of the compounds in the layer. Evidence suggests that a very active heterogeneous chemistry occurs involving marine aerosols. These reactions can have important consequences involving the loss of NO_y and other soluble gases.

5. *How does transport and chemistry in the marine boundary layer alter the pollutant mix?*
6. *How do ship emissions influence the chemistry in the marine boundary layer?*

Marine free Troposphere: Above the marine boundary layer over the North Atlantic, chemical transformation will occur in plumes that are isolated from fresh emissions. This may allow the determination of ozone and aerosol production in isolated plumes to be extended through much of the diurnal cycle.

7. *How does the rate of ozone and aerosol production and loss change during transport?*
8. *How is the chemical composition of aerosol altered during transport?*

Deployment strategy

The chemical evolution in the plumes produced by the emissions of distinct pollution source categories will be studied during the daytime and at night.

Daytime Chemistry

These studies will focus on the formation of ozone and aerosols downwind of power plants and urban plumes over regions with different natural hydrocarbon emissions.

1. *Power plant studies:* Integrated quasi Lagrangian plume studies that use the fast response measurements on board the P3 will investigate the rate and efficiency of ozone production, the rate of gas phase SO_2 oxidation (Ryerson et al., 2001, Brock et al, 2003), and aerosol formation. The studies, also, will examine the free radical chemistry as a function of the changing precursor concentrations across the plume that leads to the observed ozone and aerosol formation.

2. *Urban studies along the eastern seaboard:* The counties along the eastern seaboard from Washington, DC to Boston are ozone non-attainment areas that violate the 120 ppbv 1-hour ozone standard. The distance between Washington and Boston is 640 km. Under a wind of 5 m/s from the SW an air mass crosses this urban corridor in about 36 hours while undergoing photochemical oxidation under repeated exposure to fresh emissions of aerosol and ozone precursors during this transit. Flights across and downwind of the urban region along the eastern seaboard will study the rate and efficiency of ozone production as well as the evolution of secondary organic aerosol component.

Nighttime Chemistry

While previous airborne studies have focused on the evolution of plumes during the daytime, the approach to the nighttime measurements is relatively new and it can be anticipated that the approach will have to be adjusted throughout the study.

Systematic Plume studies will involve *in-situ* measurements of NO_2 , NO_3 , and N_2O_5 along with the end products such as HNO_3 , particulate NO_3^- , and organic nitrogen in aerosols. Aerosol formation in the reaction of monoterpenes with ozone and/or NO_3 radicals at night will be investigated.

1. *Urban:* During the late afternoon (rush hour) and at nighttime urban pollutants are emitted into the transient PBL with reduced vertical exchange. The accumulation of the pollutants in the lowest hundreds of meters renders the study of the evolution of urban emissions during the nighttime problematic. However, the experience gained during NEAQS 2002 demonstrate that nighttime emissions into the shallow boundary layer can be followed by ship borne measurements. During the 2004 study effort should be made to follow nighttime urban plumes further into the Gulf of Maine. This does not have to be done in a true Lagrangian fashion, but intermittent plume interception at various distances downwind from the sources will allow investigation of the chemical state of the plume at different ages since emission.
2. *Power Plant:*
 - a. Air borne studies of the evolution of power plant plumes will focus on the oxidation of NO_x emitted during the nighttime into the stable remnant PBL. Due to the absence of turbulent mixing at night these plumes will be concentrated in well-defined layers.
 - b. A second focus of the nighttime study will be to determine the fate of remnant NO_x that was emitted during daytime. As turbulent mixing decreases in the late afternoon vertical wind shear can contribute to the horizontal dispersion of NO_x in these plumes.

Isolated power plants in New England (e.g. Brayton, MA, Merrimack, NH) and in Pennsylvania (e.g. Montour, PA) will generate plumes suitable for the study. In addition, flights could also follow the emissions power plants located near Pittsburgh. In addition, the cluster of large power plants and industrial facilities around Pittsburgh will allow the investigation of ozone and aerosol production during transport from the Ohio River valley to the eastern seaboard. Coordination with the Lidar aircraft will make it easier to locate the height of the nocturnal plumes from these power plants.

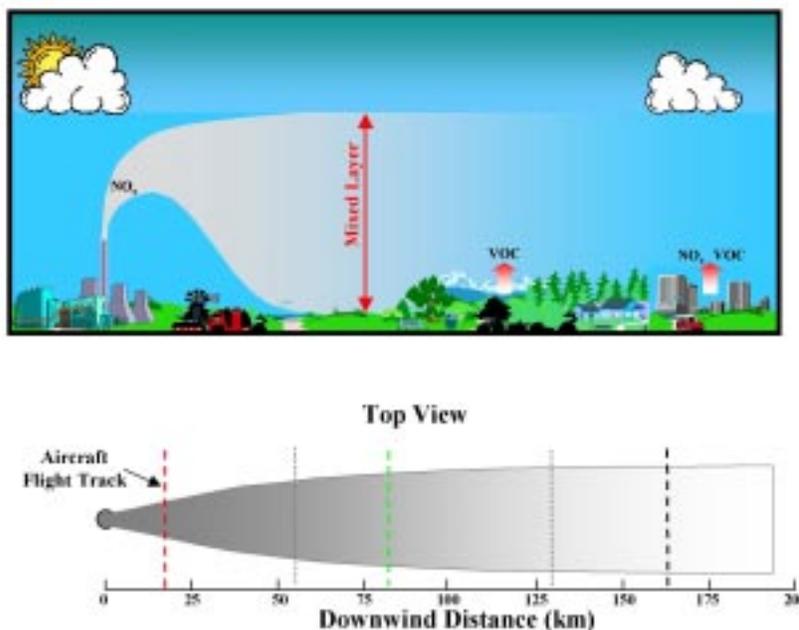


Figure 8. Schematic of the vertical (top) and horizontal (bottom) evolution of a power plant plume emitted into the daytime mixed layer. Aircraft cross-wind transects designed to study the chemical evolution of the emissions are shown in the lower part of the figure.

Chemistry during transport to the North Atlantic

These measurements will follow the formation of ozone and aerosols along with oxidation of their precursors compounds and other secondary trace gases as well as aerosols. The measurements will be used as input for constrained box models that can calculate the ozone production and loss terms.

1. The strongest signal of export to the North Atlantic can be seen ahead of frontal passages that follow the accumulation of pollution during high-pressure episode over the continent. An example of this type of investigation is the measurements that were made on August 28, 1993 during the NARE 93 campaign (Buhr et al., 1996, Daum et al., 1996).
2. When the synoptic transport carries air from the west to northwest the WP-3D can follow the chemical evolution of distinct urban plumes during transport. (An example of such a scenario is the flight on March 27, 1996.). These studies could profit greatly by being done in coordination with remote sensing aircraft that can guide the WP-3D to the center of pollution layers while at the same time determining their vertical extent.

Aerosol Properties and Radiative Effects

Relevance

Atmospheric aerosol particles affect the Earth's radiative balance directly by scattering (Charlson et al., 1992) and absorbing (Ramanathan and Vogelmann, 1997) sunlight, and indirectly by acting as cloud condensation nuclei (CCN), thereby influencing the albedo (first indirect effect, Twomey, 1991), life-time (Albrecht, 1989), precipitation (Rosenfeld, 2000) and extent (Ramanathan et al., 2001) of clouds. In many regions the natural aerosol has been substantially perturbed by anthropogenic activities, particularly by increases of sulfates, nitrates, organic condensates, soot, and soil dust. The present day global mean radiative forcing due to the direct and first indirect effects of tropospheric anthropogenic aerosol particles is estimated to be between -1.1 and -2.7 Wm^{-2} , which must be compared with the present day forcing by greenhouse gases of between $+2.2$ and $+2.6 \text{ Wm}^{-2}$ (IPCC, 2001). The geographical distribution of aerosols differs from that of greenhouse houses, however, due to shorter aerosol lifetimes (in the range of 4-5 days, *IPCC, 1994*). Aerosol concentrations are particularly high in regions downwind of sources where diurnally-averaged clear sky surface forcings range up to 30 Wm^{-2} (Russell et al., 1999; Ramanathan et al., 2001). This uneven forcing can cause continental to hemispheric scale effects on climate patterns.

International field campaigns during the past 7 years have studied aerosol properties and their direct radiative effects downwind of North America (1996 - TARFOX), Europe (1997 - ACE-2), SE Asia (1999 - INDOEX), and Asia (2001 - ACE-Asia). ACE-2 also used multiple aircraft to measure the first indirect effect of aerosols. More recently, shipboard measurements during NEAQS 2002 along the U.S. East Coast showed that the U.S. pollution plume can be as intense (in terms of aerosol mass concentration, aerosol optical depth, and ozone mixing ratio) as those downwind of India and Asia (Quinn and Bates, 2003). NEAQS 2002, however, did not specifically measure the direct or indirect radiative effects of these aerosol pollution plumes.

Science Questions

During these 7 years of major field programs our scientific tools for measuring aerosol and cloud properties and our understanding of the complex chemistry and transport processes have evolved tremendously. We propose to use these new tools to address the following questions:

Aerosol Characterization Characterizing the regional distribution of aerosol properties and the relationships between these properties are essential for assessing the direct and indirect aerosol radiative forcing and developing a predictive understanding of the climatic impact of atmospheric aerosols.

1. What are the chemical, physical and optical properties of the aerosol particles advecting from North America out over the Northwestern Atlantic Ocean?
2. How do these properties change with altitude, location, distance from the continent, time of day, and with changes in meteorological conditions?

These questions also address a key issue in air quality research: what are the main chemical constituents contributing to the regional haze? It is clear from NEAQS 2002 that particulate organic matter is a large fraction of the total aerosol off the coast of North America compared to the aerosol downwind of India (INDOEX) and Asia (ACE-Asia) (Quinn and Bates, 2003). What is the composition of this organic aerosol and what are the main sources?

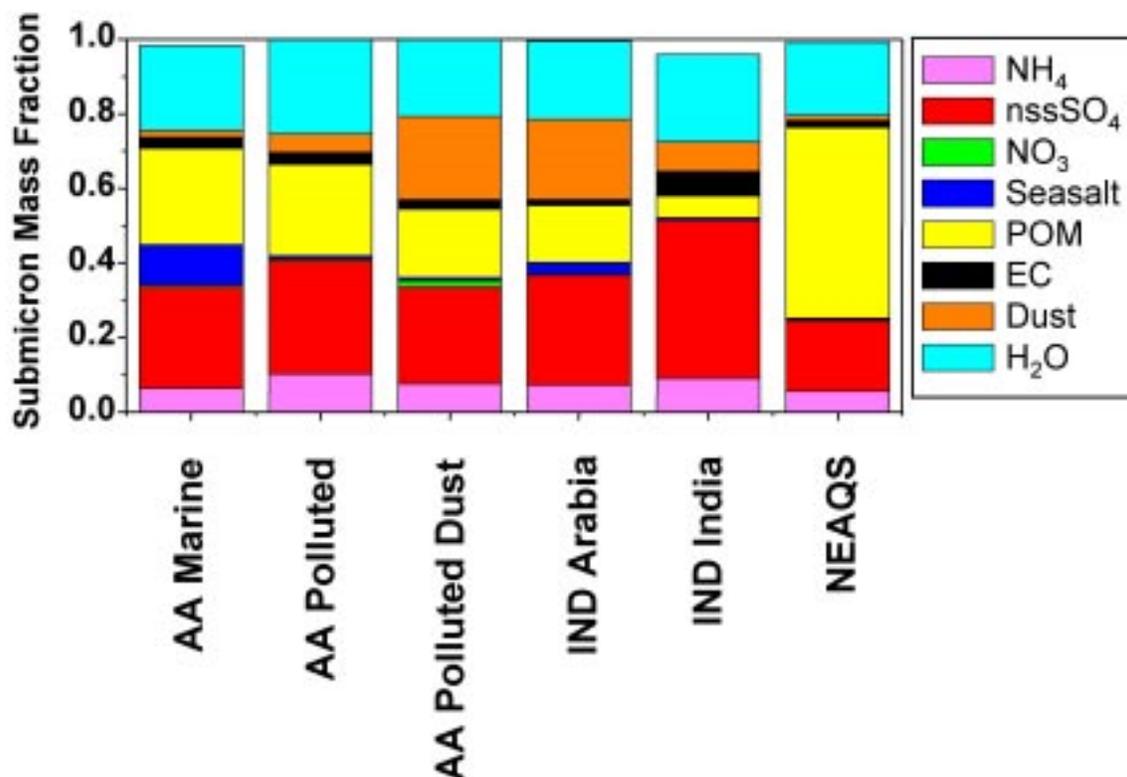


Figure 9. Average submicron mass fractions (MF) of the aerosol chemical species for ACE Asia, INDOEX, and NEAQS 2002. MF are based on the gravimetrically-determined aerosol mass and concentrations of the chemical species. The MF of H₂O is that calculated to be associated with the ionic chemical species at 55% RH. POM is estimated by multiplying the measured concentration of OC by a factor of 1.6 to 2.1 to account for associated H and O [Turpin and Lim, 2001].

Direct Radiative Effects

Relate the optical properties of aerosols to their microphysical and chemical properties and identify the processes that determine those properties.

3. What is the clear-sky radiative impact of the aerosols advecting from North America out over the Northwestern Atlantic Ocean?

In-direct Radiative Effects

The uncertainty in the "indirect effect" of aerosols on climate, i.e., in their roles in changing clouds and cloud properties, is recognized as the largest uncertainty in aerosol radiative forcing and is noted to be the largest single contributor to the uncertainty in radiative forcing by anthropogenic activities since 1750 [IPCC, 2001]. Therefore, a better characterization of the indirect effects of natural and anthropogenic aerosols is crucial for a more-accurate prediction of the impact of human activities on climate [NACIP, 2002].

4. How do the different continental aerosols advecting from North America out over the Northwestern Atlantic Ocean affect the cloud drop size distribution and cloud reflectance (first indirect effect)?

Deployment Strategy

Regional characterization and direct radiative effect

Use a combination of chemical forecast models and surface and airborne lidars to determine the location and vertical distribution of aerosol plumes. Position the ship and

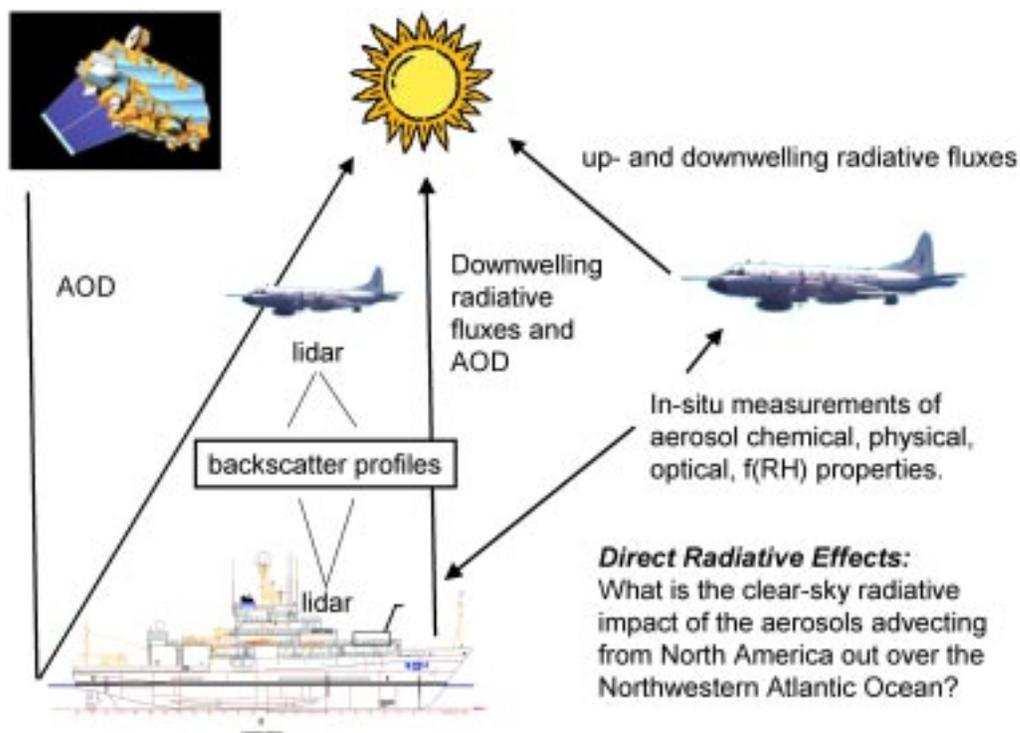


Figure 10. Quantifying the direct radiative effects.

aircraft to make *in situ* measurements of aerosol size distributions, chemical composition, $f(\text{RH})$ and $g(\text{RH})$, and scattering and absorbing properties within the plumes at the surface (ship) and aloft (aircraft). Based on these measured aerosol properties, calculate the clear sky forcing for different regions, aerosol sources, and meteorological conditions. In addition, measure downwelling radiative fluxes at the surface (ship) and up- and downwelling radiative fluxes of aerosol layers aloft (aircraft). Measure surface (ship) and top of atmosphere (satellite) aerosol optical depth. Compare the surface forcing derived from the flux radiometers with those derived from models that use the *in situ* measurements as input parameters.

Measurements: To determine the aerosol clear sky radiative forcing a combination of *in situ* measurements and passive and active remote sensors onboard ship and aircraft platforms is required. *In situ* measurements will be focused on an accurate determination of the aerosol size distribution, chemical composition, and scattering, backscattering, absorption and extinction coefficients (ship and aircraft). Lidars will be used to determine the height, depth, and homogeneity of aerosol layers (ship and aircraft) as well as the horizontal extent and homogeneity (ship). Flux radiometers will be used determine upwelling and downwelling fluxes and to derive surface and top of atmosphere forcing (ship, aircraft, and satellites). Sunphotometers will be used to measure aerosol optical depth (ship and satellite). Combining these measurements will allow for a complete characterization of the aerosol at the surface and throughout the lower troposphere and a determination of clear sky forcing through a variety of techniques.

In situ measurements:

- Aerosol microphysics: Total number concentration and size distributed number concentration (3 nm to 10 μm).
- Aerosol chemical composition: Size segregated aerosol mass and concentration of inorganic ions, total organic carbon, total elemental carbon, speciated organics, and trace elements.
- Aerosol optical properties: Scattering, hemispheric backscattering, absorption, and extinction coefficients of the sub-1 and sub-10 μm aerosol at three wavelengths (450, 550, and 700 nm).
- Aerosol light scattering hygroscopic growth ($f(\text{RH})$) and aerosol size hygroscopic growth ($g(\text{RH})$).

Remote measurements:

- Vertical profile and horizontal extent of aerosol backscatter
- Aerosol optical depth at 380, 440, 500, 675 and 870 nm
- Clear sky radiative fluxes (spectral? Broadband? Wavelength range?)

Verification/Intercomparison:

The following represent a set of planned experiments to intercompare and/or verify critical measurements:

- Comparison of directly measured aerosol scattering, backscattering, and absorption coefficients with those calculated from the measured size distributions and chemical composition (local closure).

- Comparison of directly measured $f(\text{RH})$ and $g(\text{RH})$ with values calculated from aerosol size distributions and chemical composition.
- Comparison of measured aerosol properties with those determined from chemical transport models.
- Comparison of aerosol extinction at the surface measured by several techniques
- Comparison of aerosol optical depth and radiative fluxes determined from shipboard, aircraft, and satellite sensors (clear sky column closure)
- Comparison of clear sky forcings derived from models using measured aerosol properties and flux radiometers

Indirect radiative effect

Measure in-situ aerosol properties below cloud (ship and aircraft); measure lidar backscatter in updrafts below cloud (i.e., aerosol entering cloud); measure liquid water path, cloud optical depth, cloud drop size using ship-based remote sensing, measure cloud drop size distribution and liquid water content in cloud (aircraft), measure cloud reflectance, optical depth and effective radius above cloud (aircraft).

Measurements: NEAQS-ITCT 2004 represents an opportunity to demonstrate capabilities of measuring important components pertaining to the aerosol indirect effect and demonstrating the capability to detect and quantify the first aerosol indirect effect in subsequent years. To maximize the value of the data from platforms such as the WP-3D and *Ronald H. Brown*, which might not be collocated on a regular basis, it is proposed that each platform be equipped with a set of instruments that will allow independent measurement of the components required to address the first indirect effect - i.e., aerosol, drop size, and liquid water. On the WP-3, this complement should include measurements of aerosol size distribution/ composition, in-situ cloud probes, and remote measurement of cloud reflectance, cloud optical depth and drop size (downward looking spectral radiometer). When flying below cloud, upward looking radiometers can also provide information on cloud optical depth, liquid water path and drop size. Statistical analyses of the relationship between aerosol parameters and cloud response will be performed. Similarly, on *Ronald H. Brown*, the combination of surface aerosol size distribution/composition, lidar backscatter, radar or radiometer-derived drop size, and liquid water path will provide another set of independent measurements. During WP-3 overflights of *Ronald H. Brown* redundancy will provide important information for intercomparison and verification of measurements, as described below. Overflights will also enable a column-oriented study if the boundary layer is convective.

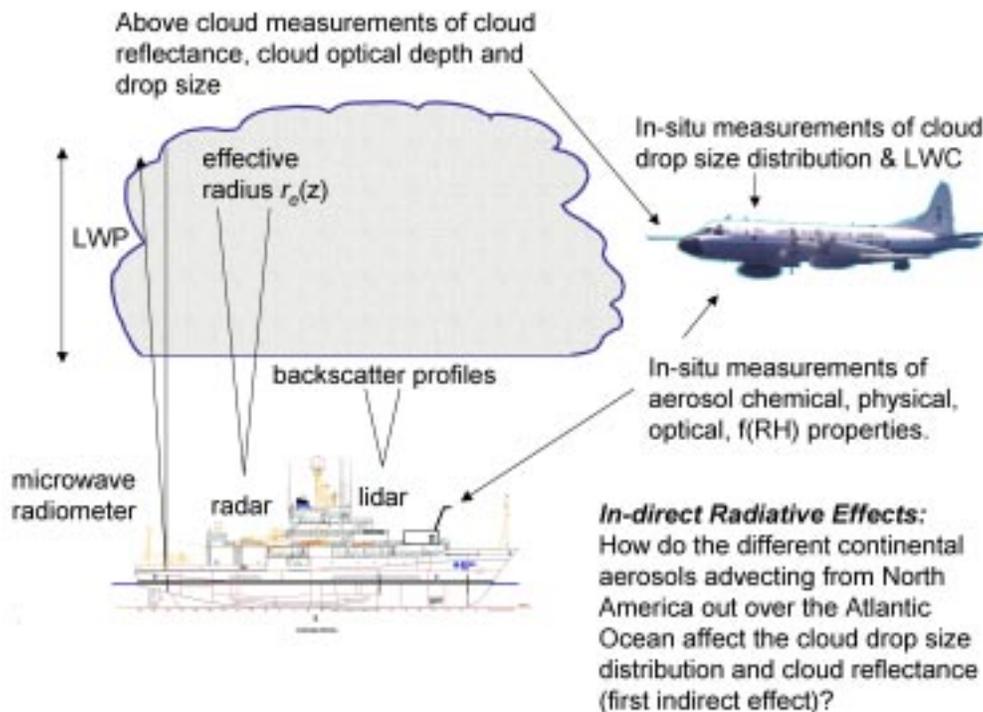


Figure 11. Quantifying the in-direct radiative effects.

Verification/Intercomparison

The following represent a set of planned experiments to intercompare and/or verify critical measurements:

- Comparison between *in-situ* size distribution and composition from the WP-3D, and surface aerosol size distribution and composition when the boundary layer is well-mixed;
- Comparison between aerosol backscatter based on WP-3D size distribution/composition and lidar backscatter;
- Comparison between $f(RH)$ from *in-situ* size distribution/composition, humidified nephelometer, and from lidar (vertically pointing, when the boundary layer is well-mixed, using clouds as calibration targets);
- Comparison between backscatter based on surface aerosol and lidar backscatter (horizontal beam);
- Comparison between radar-derived drop effective radius r_e and ship-board spectral measurements;
- Verification of remote r_e measurements by comparing radar or radiometer-derived r_e with WP-3D *in-situ* measured r_e (FSSP probe);
- Comparison between drop size distributions from WP-3D FSSP probe and Twin Otter FSSP and/or new phase-Doppler instrument;
- Comparison between downward looking WP-3D-based retrievals of cloud optical depth, r_e and liquid water path with similar retrievals from ship-based remote sensors (radar, radiometer, lidar, microwave radiometer);

- Comparison of liquid water path from aircraft profiles and ship-based microwave radiometer in the event of horizontally homogeneous conditions.

Measurements and Modeling: The measurements to be acquired during NEAQS-ITCT 2004 represent an opportunity to test the ability of cloud parcel models to represent cloud droplet formation. Models will use size distribution /composition, and vertical velocity measurements (from aircraft, Doppler lidar, or Doppler radar) as input. Drop size distributions (particularly droplet concentration) will be compared with observed values. Comparisons will be made in a statistical sense by using probability density functions (pdfs) of input parameters and comparing pdfs of model output with pdfs of observed parameters. In addition, a large eddy simulation model that integrates coupled dynamics, aerosol and cloud microphysics, aqueous chemistry and radiation, will be applied as an interpretive tool to a number of case studies.

Forecast Models

Relevance

The effective management of the Nation's air quality requires predictive capabilities as demonstrated in a reliable air quality forecasting system. The envisioned system would provide industry and local and state agencies with forecasts that indicate when and where incidences of reduced air quality are going to occur. NOAA has undertaken an initiative program that will lead to improved air quality forecasting in the United States. The system would combine an adequate understanding of the basic chemical and dynamical processes that determine atmospheric composition, reliable emission inventories, dependable forecast models and an adequate monitoring network that provides information needed to initialize the models.

An essential component of the system is the development of adequate model systems that synthesize our current understanding of atmospheric transport, emissions, chemical, and physical transformations of key pollutants and their precursors. During the 2004 field study, NOAA will deploy two air quality forecast models that can provide operational air quality forecasts. The evaluation of these models with data collected in the 2004 field study will allow model developers valuable insight concerning the applicability of the various model components that comprise the air quality forecast model.

In addition to goals aimed at development of operational air-quality forecasting models the study in 2004 will allow informal comparison of forecast made by regional air-quality models and chemical/transport models that operate at hemispheric and global scales. The information will allow conclusions to be drawn concerning the reliability of the different model simulations as a function of altitude and proximity to major sources.

Science questions

Air quality forecast models are essentially a computational synthesis of our collective understanding of how anthropogenic pollutants are emitted, transformed, and transported. The various measurements and platforms that constitute the NEAQS-ITCT 2004 field study will provide diverse, and rigorous tests to this basic understanding. A model-measurement comparison should not only characterize the accuracy of the forecast models but also identify elements of the models that limit their accuracy, and point the way to better forecasts. On the other hand utilizing the predicted meteorological and chemical fields from forecast models is also an important component of a well-integrated field campaign. There is also an important time interval, during the first examination of a day's measurements, when it is extremely valuable to principal investigators and experiment planners to know what the forecast models predicted. These inherent

synergisms between the field experiment and the forecast models are the impetus and framework for the forecast component of the NEAQS-ITCT 2004 study.

1. *How well can air quality models forecast air quality in New England?* One of the important goals of the NEAQS-ITCT 2004 air quality study will be to determine how well current state-of-the-art air quality models can forecast air quality in New England. There is a twenty-year precedence for the statistical evaluation of ozone predicted by air quality models based primarily on comparisons with the EPA AIRS air quality monitoring network. Model evaluation studies for aerosols and the precursors for aerosol and ozone are severely limited by a lack of data both aloft and at the surface. The 2004 study is unique in that it will provide a glimpse of both the gas-phase oxidant component of air quality (i.e. ozone) and the particulate-phase components (i.e. PM_{2.5} and PM₁₀ aerosol) over a large region and altitude extent of the Northeast U.S. This data set will therefore represent the centerpiece not only for model evaluation of ozone and its precursors, but for aerosols and visibility in the Northeast U.S. as well.

Model evaluations are obviously most meaningful when results from two or more independent models are available for coincident comparisons. The cross-evaluation of several air quality forecasts is an important aspect of the NEAQS-ITCT 2004 evaluation study. There are fundamental differences between the CMAQ-ETA (off-line meteorology drives pollutant transport) and the WRF-CHEM model (online, or lock-step calculation of meteorology and pollution transport) that justify a detailed statistical evaluation between these two models. Other important elements to air quality forecasts, such as the treatment of vertical transport and turbulent mixing, the photochemical mechanism, and the sensitivity to horizontal resolution can only be compared and evaluated within the context of multiple model forecasts.

2. *How accurately do the forecast models represent the individual processes controlling air pollution formation and transport?* While the first science question addresses the end result of pollution formation, and the raw output of the model forecasts, it is important from a scientific perspective to determine how accurately the forecast models represent the individual processes controlling air pollution formation and transport. Three broad subsets of processes are the focus the NEAQS-ITCT 2004 field program.

Emissions estimates: Air quality forecast models are fundamentally limited by the accuracy of the emissions estimates of ozone and aerosol precursors imposed on the model. Conversely, the considerable resources and effort the U.S. EPA puts into quantifying emissions from thousands of U.S. sources, and eventually deriving an emissions inventory, is not sufficiently matched by resources or effort into validation of the inventories. As discussed in the emissions evaluation section of this report, a key focus of the aircraft and ship-based platforms during NEAQS-ITCT 2004 is the evaluation of emissions inventories, on a relative as well as an absolute basis. Air quality forecast models are an important,

computational intermediate that relate the emissions inventories to atmospheric concentrations. The model evaluation study will provide the developers of the forecast models a clear picture on the ability of the models to capture both relative and absolute precursor abundances, allowing indirect evaluations of the magnitude and relative location of the sources in the emissions inventories.

Photochemical and physical transformations: Since ozone, and particulates to a large degree, are secondary products formed during the oxidation and transport of primary emitted species, the accuracy of air quality forecasts is highly dependent on the veracity of the model's various transformation processes. The quasi-Lagrangian aircraft and ship-based studies of individual sources and urban regions planned for NEAQS-ITCT 2004 lend themselves directly to the evaluation of the photochemical and/or aerosol mechanisms within each forecast model. The forecast models predict key oxidants (OH, O₃ in the daytime, NO₃ and N₂O₅ at night), as well as the various secondary products produced from primary nitrogen, sulfur, anthropogenic and biogenic hydrocarbon emissions that will be measured during the field study. Comparisons between observed and modeled relationships among the various secondary and primary emitted species in combination with oxidant abundances will allow both quantitative and relative evaluations of the individual forecast models.

Meteorology and transport: The experience from the previous research outlined in the beginning of this report has unequivocally shown that the key to understanding pollution at the surface is to understand the processes controlling pollution aloft. The various upper-air platforms within the NEAQS-ITCT 2004 field program (aircraft, wind profilers, ozone and aerosol lidars, doppler lidar, ozonesondes and radiosondes) provide broad coverage in terms of area, physical, and photochemical parameters with which to evaluate the forecast models. Since forecasting winds, convection, and vertical transport correctly are prerequisite to accurate air quality forecasts, it is particularly important to evaluate the model's ability to adequately characterize the various scales of transport, from near surface to synoptic and regional scales.

Deployment Strategy

Lessons Learned in 2002

The air quality forecast model evaluation component of the NEAQS 2002 pilot study serves as a useful framework for model evaluation planning and deployment during NEAQS-ITCT 2004. Two models with significant differences in terms of their basic structure, physical parameterizations, photochemical mechanisms, and emissions processing were evaluated. The model forecasts were compared with measurements of ozone and its precursors that were taken at the 4 University of New Hampshire AIRMAP ground sites, the Harvard Forest site, and aboard *Ronald H. Brown* research vessel. Time series of preliminary observations and model forecasts of O₃, CO, reactive nitrogen, SO₂,

sulfate, and a number of meteorological parameters were posted on a web site in near real-time that was accessible to the planners, participants and forecasters involved with the field program, allowing a qualitative glimpse of forecast reliability relative to the observations. Model forecasts were also used to direct the deployment of *Ronald H. Brown*. This allowed the ship to intercept urban plumes during the study period that proved useful for model evaluation.

Close collaboration between the individual air quality forecast groups and evaluation team is essential to the success of a formal evaluation study. A pre-deployment consensus on evaluation protocol, model domains, common data sets and model products will be necessary. One of the lessons from the NEAQS 2002 pilot study is that a large uncertainty in explaining model differences would be eliminated if the emissions inventory were consistent between the models. It is highly recommended that a common emissions inventory for seven ozone and particulate precursors (NO_x, CO, VOC, SO₂, PM₁₀, PM_{2.5}, and NH₃) be made available to those forecast and modeling groups involved in NEAQS-ITCT 2004 to insure compatibility between different models and different model resolutions. A “reference standard” inventory with sufficient temporal and spatial information would benefit not only the NEAQS-ITCT 2004 study, but the air quality community in general, much as the OTAG and NAPAP inventories have done previously.

Regional Air Quality Forecast Models

The NEAQS-ITCT 2004 study will involve a real-time comparison phase based on the preliminary field measurements, a post-field study phase where evaluations are performed for the entire summer based on the preliminary measurements, and a final evaluation phase based on finalized, quality assured data. It is anticipated that models will be run during 2004 to evaluate skill at predicting ozone photochemistry and PM₁₀ and PM_{2.5} aerosol concentration. Forecasts will be available at several model resolutions. Nested domains of 4 and 12 km centered over the northeast U.S. within a 36 km grid covering the entire U.S. for the WRF-CHEM model are currently being considered. The two regional-scale forecast models (CMAQ-ETA and WRF-

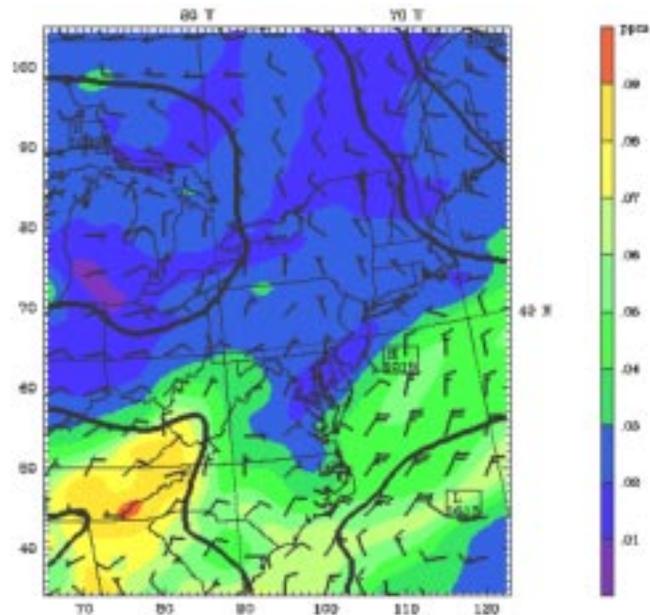


Figure 12. 12-hour WRF-CHEM prediction for July 12, 00Z, 2002

CHEM) are the top priorities in terms of model evaluation studies due to their spatial detail, the effort and

expense put into the basic physics and dynamics, and the importance they serve as operational or community based forecast models.

Additional Air Quality Models

A number of models that participated in ITCT 2002 are expected to be operational during NEAQS-ITCT 2004. These include the Harvard University GEOS-CHEM model, the MOZART model from GFDL/NOAA, the University of Iowa long-range transport model, and the FLEXPART trajectory based model of Andreas Stohl. The coarse horizontal resolution and scale of these models are best suited for intercontinental transport studies, but they naturally include the Northeast U.S. as a source region. These models will therefore provide additional points of comparison for the observational platforms, albeit without the detail of the regional AQ forecast models. Hopefully, the groups in charge of these models will undertake their own, independent comparisons of model results with observations from the NEAQS-ITCT 2004 research communities similar to the activities during ITCT 2002.

Model Evaluation

During NEAQS 2002, real-time comparisons of model forecasts were provided to ground-based and ship-based observations. Plans are being made to provide similar forecasts during 2004 to facilitate model evaluation. In addition, comparisons of model forecast results with the aircraft platform(s), and comparisons of model forecast aerosol fields with the observations will be undertaken. The software, hardware, and personnel are already positioned to assume these additional tasks. There are five advantages to having the Aeronomy Lab as the central contact in the real-time model evaluation study: 1) most of the real-time air quality and meteorological data collected during the field study will reside on Aeronomy Lab computers, 2) allows the forecasting groups to avoid duplicate efforts of model/observation comparisons, freeing up time and resources, 3) provides a central location for real-time comparisons available to mission planners, model forecasters and field scientists, 4) provides a repository for model forecasts that can be used in more detailed analysis after completion of the study, 5) the infrastructure for rapid dissemination of model/observation comparisons is already in place at the Aeronomy Lab.

Post-Deployment Strategy

The post-analysis of model forecasts is scientifically the most important aspect of the evaluation study. Only through a quantitative, statistical framework with sufficient sampling throughout the entire ozone season can relative model performance be judged fairly. Having the air quality forecasts in a central location will be very convenient for the post-analysis that will need to be undertaken.

Mobile Platform Descriptions

NOAA Research Vessel *Ronald H. Brown*

Transport of polluted air within the Gulf of Maine plays an important role in shaping the air quality in coastal New England. These same pollutants can affect the regional radiation budget as well as precipitation and the lifetime and extent of clouds. The polluted air is a result of both re-circulation of pollution from urban areas within New England and long-range transport (e.g., from populated areas along the eastern seaboard and the Ohio River Valley). Over the Gulf of Maine the marine boundary layer (MBL) can act as a huge chemical reactor converting primary pollutants like nitrogen oxides and organics into more-toxic secondary pollutants like ozone and fine particles which can be transported back onto shore in the land-sea breeze circulation.

An instrumented ship is an ideal platform to study the meteorological and chemical



Figure 13. The NOAA Research Vessel *Ronald H. Brown*

processes that are occurring off the coast of New England. A ship can be used to sample polluted air masses as they move offshore, study the chemical transformations in the polluted marine boundary layer, and characterize polluted air masses as they move onshore. Indeed, deployment of *Ronald H. Brown* during NEAQS 2002 demonstrated unequivocally the value of this platform for providing unique sampling opportunities, unlike those from sites on shore that result in data that are difficult to interpret due to contamination by local land-based sources and unlike those from aircraft that have short duration and result in limited data sets.

Ship Capabilities and Facilities

The operational capabilities and shipboard facilities of *Ronald H. Brown* are shown in Table 1 (see also <http://www.pmc.noaa.gov/rb/>). The instrumentation (payload) capacity

Table 1. Performance specifications and facilities for *Ronald H. Brown*.

Parameter	Specification
Length (ft/m)	274 / 83.5
Range (nm/km)	11,300 / 20,900
Endurance (days)	35
Cruising speed (kts / mps)	12 / 6.2
Maximum speed (kts / mps)	15 / 7.7
Officers / Engineers / Crew	5 / 4 / 16
Scientific staff	34 (maximum)
Laboratory/office space (sq. ft.)	4100
Telecommunications, data	INMARSAT-A
Telecommunications, voice	Cell & satellite phones, VHF radios

of *Ronald H. Brown* is not limited by weight or power constraints. Typically, atmospheric sampling instruments are placed in seagoing laboratories (sea-tainers) on the forward upper (02) deck (see Figure 13). Air samples are collected using towers or masts that extend 6-8 meters above the deck (approximately 16 - 18 meters above the water line). Sampling is conducted around the clock, unless contamination from the ship exhaust is expected to be prolonged. Remote sensing meteorological measurements are also included in the instrument package to define the structure and extent of the MBL and thus place the chemical measurements in context. Augmentation of the on-board radar wind profiler with additional lidar instruments is critical for this activity.

The ship is capable of staying out to sea for long periods, which allows for repeated sampling of air masses in a particular region, such as the Gulf of Maine. However, with an average cruising speed of 12 knots the ship is not a rapidly moving platform. For example, at the nominal cruise speed (22 km hr^{-1}) the 400 km trip from Boston, MA, to Yarmouth, NS, takes 18 hours. Within certain constraints *Ronald H. Brown* is capable of extended near-shore running, which is especially valuable for examination of pollution plumes advected off the shore and for examination of meteorological phenomena such as land-sea breeze effects. The ship is fully capable of nighttime operations, though with some restrictions when near shore.

Instrumentation

Table 2. Proposed instrumentation for deployment on *Ronald H. Brown*.

Parameter	Method
Photolysis rates (j-values)	Spectral radiometer
Ozone (O ₃)	UV absorbance
Ozone	NO chemiluminescence
Carbon monoxide (CO)	Nondispersive IR
Carbon dioxide (CO ₂)	Nondispersive IR
Sulfur dioxide (SO ₂)	Pulsed UV fluorescence
Nitric oxide (NO)	Chemiluminescence
Nitrogen dioxide (NO ₂)	Photolysis/chemiluminescence
Total reactive nitrogen oxides (NO _y)	Au tube/chemiluminescence
Peroxyacyl nitric anhydrides (PANs)	GC/ECD
Alkyl nitrates (RONO ₂)	GC/MS
Nitrate radical (NO ₃); Dinitrogen pentoxide (N ₂ O ₅)	Cavity ring-down spectrometry
Nitric acid (HNO ₃)	Mist chamber/IC
Water vapor (H ₂ O)	Nondispersive IR
Continuous Speciation of VOCs	PTR-MS/CIMS
VOC Speciation	GC/MS
Formaldehyde (HCHO)	CHD fluorimetry
Radon (Rn)	Radon gas decay
Seawater/atmospheric CO ₂	Nondispersive IR
Enhanced measurement of radiative fluxes	Spectral radiometers
Aerosol optical depth	Continuous sunphotometer
Aerosol optical depth	MicroTOPS
Irradiance	Portable Radiation Package (PRP)
Size-resolved aerosol composition and gravimetric mass	Impactors (IC, XRF, and thermal-optical OC/EC)
OC/EC	On-line thermal optical
Ionic Aerosol Composition	Particle In Liquid Sampler (PILS)
Aerosol Size and Composition	Aerosol Mass Spectrometer
Aerosol scattering (400, 550, 700 nm)	TSI Model 3563 Nephelometer
Aerosol absorption (550 nm)	Radiance Research PSAP
Aerosol number	CNC
Aerosol size distribution	Twin DMAs and an APS
Aerosol light scattering hygroscopic growth f(RH)	Twin TSI 3563 nephelometers
Aerosol size hygroscopic growth g(RH)	Tandem DMAs
Total and sub-micron aerosol extinction	Extinction cell
Total and sub-micron aerosol extinction	Cavity ring-down spectrometer
Ozone/aerosol vertical profiles	O ₃ /Aerosol Lidar (OPAL)
Wind/temperature vertical profiles	915 MHz wind Radar
High-resolution BL winds/aerosol	Doppler Lidar (HRDL)
Wind profiles/microscale turbulence	C-band radar
Temperature/relative humidity profiles	Radiosondes
Surface energy balance (fluxes)	Eddy covariance (bow mounted)
High resolution BL turbulence structure	Doppler mini-Sodar
Cloud drop effective radius	K-band radar
Cloud liquid water path (aerosol indirect effect)	Microwave radiometer
Liquid water path	Mini Differential Absorption Spectrometer (MIDAS)

The instrumentation proposed for deployment on RHB (see Table 2) will provide characterization of the atmospheric dynamics, gas-phase chemistry, aerosol chemical, physical, and optical properties, and radiation fields in this complex environment. Central to the NEAQS-ITCT 2004 will be techniques that yield information on the interactions between gas-phase and aerosol chemistry and how the evolving aerosol properties affect the radiation fields. A critical requirement is understanding how these chemical effects are influenced by transport to, from and within the MBL. Thus particular emphasis has been placed on the need to understand the dynamical structure of the MBL at large scales via remote sensing instruments. Smaller scales will also be studied with the addition of instrumentation to investigate the turbulence structure from the surface layer to the top of the MBL. Coincident with this activity will be measurements of chemical fluxes of CO₂, and possibly O₃.

Ship Operations

Because the ship cannot rapidly deploy to different areas to take advantage of sampling opportunities, meteorological forecasting is essential for planning ship operations. Coordination of these forecasts (meteorological and air quality) with ship track planning in 2002 was very successful. This activity will be augmented in 2004 by more frequent communication between the ship and forecast personnel on shore. In addition for the NEAQS-ITCT 2004, extensive coordination between the ship and the various aircraft will be required. Coordination with the airborne lidar will be especially beneficial for the ship because that aircraft will be most useful for providing information on plume locations around the Gulf of Maine. Efforts will be made to maximize opportunities for measurements comparisons between the ship and the various aircraft.

A significant restriction on ship operations, and therefore ship track planning, is the need for the relative wind to be forward of the beam of the ship in order to avoid sample contamination from the ship exhaust. Accurate forecasting of surface winds is essential for this; just as essential is having several sampling options available (see below). Since this strategy worked very well during the NEAQS 2002, it will be expanded upon for 2004.

Ship Sampling Objectives

Using the experience gained in 2002, the sampling tracks for Brown in 2004 will be planned 1) to examine further the processes that influence pollution episodes along the coast of New England and 2) to examine the effects of aerosol transport and transformation on the radiative properties in the atmosphere (clear sky and cloud). The focus will be on four major scientific objectives, each associated with unique, but necessarily overlapping, sampling strategies.

Characterization of sources – Near-shore survey tracks are planned under conditions when polluted continental air is expected to be transported into the surface marine layer (i.e. nighttime, early morning, or late day). Similar tracks, likely farther north, are planned to examine the influence of biogenic emissions on photochemistry and aerosol

formation and growth in the MBL. Also, special effort will be made to characterize marine vessel emissions (MVE), probably near harbor areas such as Boston or New York.

Study of transport and transformation processes - A focus of the proposed research is the study of the evolution of polluted air masses in the Gulf of Maine. When possible, plumes advected off-shore will be sampled at successively longer distances downwind to examine chemical transformations related to plume aging in the MBL (see Figure 14). Opportunities to sample well-aged plumes that have remained in the MBL for several days should be possible with close coordination with the airborne lidar. Also, since significant chemical transformations occur at night, these studies of polluted air masses will occur during the entire diurnal cycle.

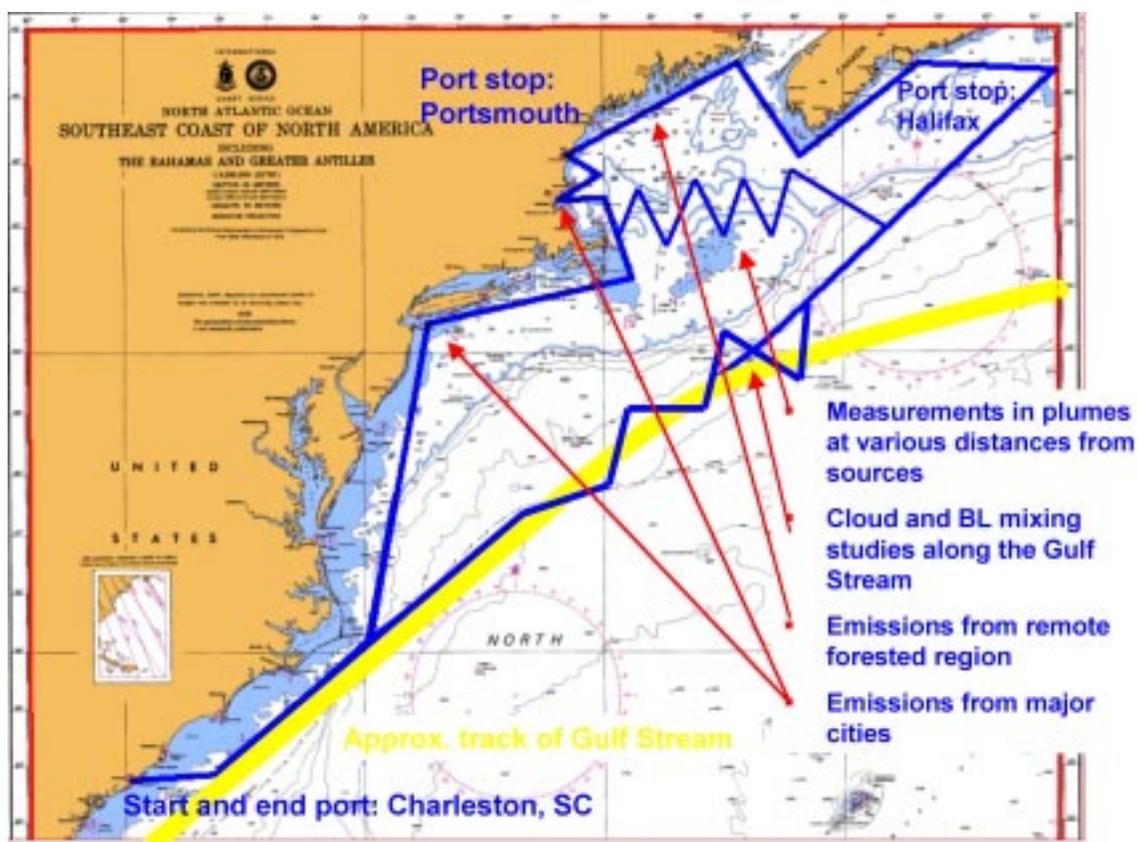


Figure 14. Possible cruise tracks to study pollution plumes and cloud effects in the MBL.

Study of coastal impacts – Air quality measurements on Brown will take advantage of the heavily instrumented region along the New Hampshire coast. Along-shore cruise tracks are planned to characterize the effects of recirculation of air masses by the sea-breeze/land-breeze circuit. More northerly tracks will be undertaken to examine the impact of long-range pollution transport in the MBL to relatively pristine areas such as Acadia National Park in Maine.

Study of radiative effects of aerosols – Cruise tracks are planned to examine the composition and structure of aerosols in both polluted (e.g., Massachusetts or New Hampshire coast) and clean (e.g., Maine or Canadian coast) conditions. Continuous measurements of radiation fields will be conducted to examine the influence of the changing MBL aerosol. An effort will be made to examine the effects of clouds on aerosol evolution. Coordination with aircraft activities will be undertaken to capitalize on opportunities to study aerosol indirect effects on radiative fields. These latter two activities are likely to require cruise tracks to the northern reaches of the study area or in the vicinity of the Gulf Stream.

NOAA WP-3D Lockheed Orion

High pollution/haze events frequently impact New England in the summer. The sources and all the factors that shape the air quality in this area are not well known. However, both local and distant sources (transported pollution) are believed to play a role. Air Pollution from the region can be advected over the pristine North-Western Atlantic with impacts on the regional and hemispheric air quality and climate.

An instrumented aircraft can uniquely address both components, e.g.:

- From the perspective of regional air-quality research, aircraft measurements can characterize an important feature of New England air quality, namely, the long-range transport of pollution from urban sources along the East Coast and from industrial regions in the Ohio River Valley. This study will address for the first time the role that nighttime chemistry and transport involving emissions from these distant sources play in shaping New England air quality.
- From the perspective of climate research, an aircraft can undertake a systematic study of the formation and evolution of the chemical and radiative properties of aerosols from urban and industrial sources as they are carried off the East Coast of the United States into the North Atlantic. The aim of such research is to address one of the more important open questions in climate research, namely, to determine how the various types of emissions and the subsequent atmospheric chemistry determine the radiative properties of aerosols and, hence, the impact of these aerosols on radiative forcing in the atmosphere.



Figure 15: NOAA Lockheed WP-3D Orion

NOAA’s Aircraft Operations Center at MacDill AFB, FL maintains and operates NOAA’s aircraft assets. Among them are two Lockheed WP-3D Orion, four engine turbo-prop aircraft, which are mainly used for severe storms and other weather related research. Starting in 1994, these aircraft have been temporarily converted into highly sophisticated airborne air chemistry and aerosol research platforms.

Platform

The operational characteristics and specifications of the NOAA WP-3D Orion aircraft are summarized in the Table 3.

Table 3: NOAA WP-3D Orion specifications and operational parameters

Parameter	Specification
Length	116’ 10”
Span	99’ 8”
Fuselage Diameter	11’ 3”
Ceiling	25,000 ft
Research Speed	200 knts IAS
Range	1600+ nm
Fuel Burn	4500 - 6000 lbs / hr
Fuel Load	58,000 lbs (48,000 lbs usable; 10,000 lbs reserve)
Max Science Payload	~ 5000 lbs inside fuselage; plus additional instruments in external wing stores

The operating range will suffice to investigate the suspected primary pollution source regions in the Ohio Valley region and along the eastern seaboard and follow the transport and transformation of their emissions across New England toward the North Atlantic (Figure 16). Figure 16 shows the range of the WP-3D operating out of Pease International Tradeport Airport for two different scenarios of 800 and 1000 nautical miles action radius, that is assuming a flight with return to Pease. The above cited operational range is an estimate and actual range will finally be determined by how much fuel can be loaded within the maximum aircraft gross weight limit of 135,000 lbs. We anticipate the aircraft to be ‘max zero fuel weight’ limited, that means the fuselage is loaded to capacity and additional instrumentation is operated in external stores (pods) under the wings.

The WP-3D aircraft are operated by an AOC crew of seven (aircraft commander, pilot, flight engineer, navigator, flight director who is also the flight meteorologist, and two technicians) and can carry in addition several science personnel. By minimizing this number we can install more instruments. The planned payload relies on the full space and payload weight capacity.

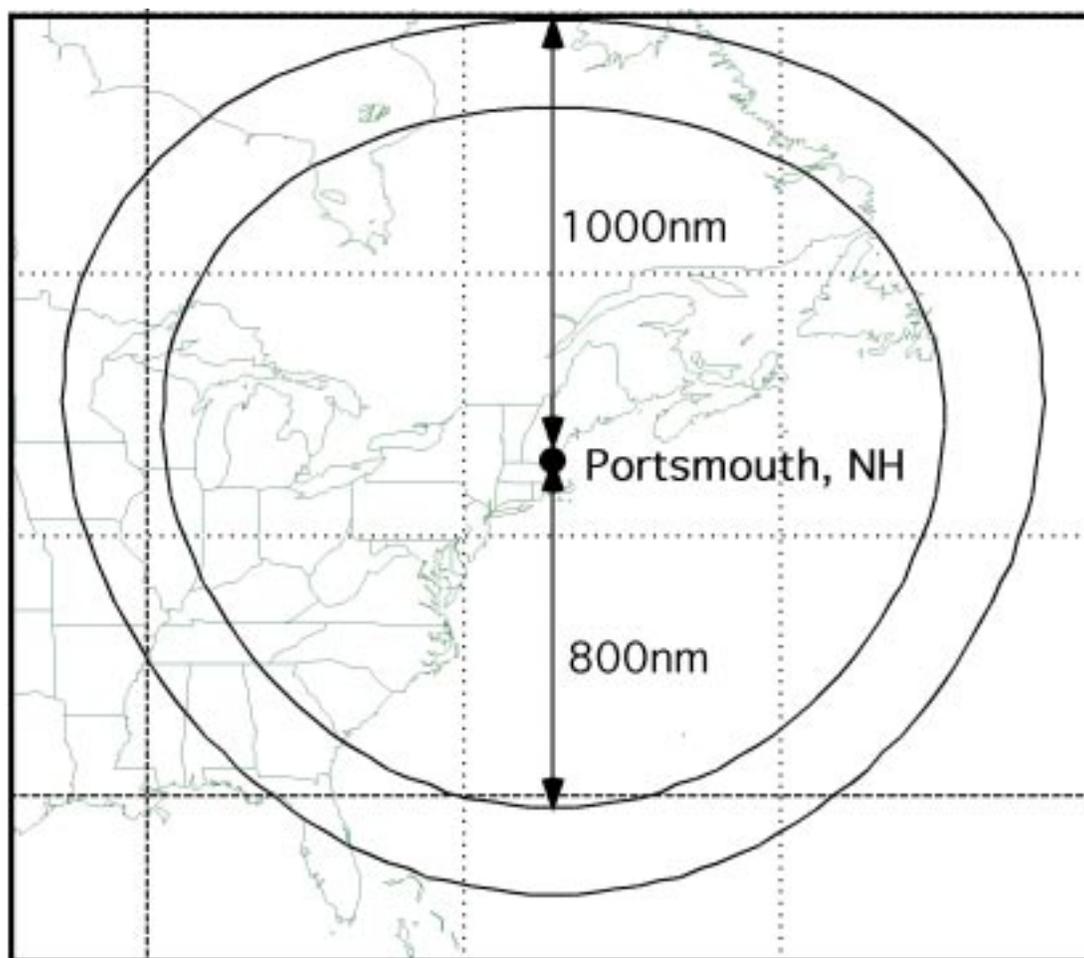


Figure 16: Operational range for WP-3D stationed at Pease International Tradeport, NH.

Instrumentation

The science instrument payload is geared to supply information on the science questions discussed above by studying daytime photochemistry, nighttime chemistry, aerosol composition and size distribution, the driving radiation fluxes, transport and chemical conversion. Details of the proposed payload are listed in Table 4.

Table 4: Proposed scientific payload for NOAA WP-3D Orion aircraft.

Parameter	Method
Ozone (O ₃)	NO/O ₃ Chemiluminescence
Nitric Oxide (NO)	NO/O ₃ Chemiluminescence
Nitrogen Dioxide (NO ₂)	Photolysis & NO/O ₃ Chemiluminescence
<i>In-situ</i> volatile organic compounds, VOCs	Proton Transfer Reaction Mass Spectrometer (PTR-MS)
Canister VOCs	Canister Sampling, GC/FID, GC-MS
Total Nitrogen Oxides (NO _y)	Au Converter & NO/O ₃ Chemiluminescence
Carbon Dioxide (CO ₂)	Non-Dispersive InfraRed (NDIR)
Sulfur Dioxide (SO ₂)	UV Pulsed Fluorescence
Carbon Monoxide (CO)	VUV Resonance Fluorescence
Formaldehyde (CH ₂ O)	Tunable Diode Laser Absorption Spectrometry
PAN's (PAN, PPN)	Chemical Ionization Mass Spectrometer (CIMS)
HNO ₃ , NH ₃	CIMS
Hydroxyl Radical (OH)	CIMS
Sulfuric Acid (H ₂ SO ₄)	CIMS
NO ₃ , N ₂ O ₅	Cavity Ring-Down Spectroscopy (CARDS)
Aerosol Single Particle Composition	Particle Analysis by Laser Mass Spectrometry (PALMS)
Aerosol Bulk Ionic Composition	Particle Into Liquid Sampling (PILS)
Aerosol Bulk Composition	Aerosol Mass Spectrometer (AMS)
Small Aerosol Concentration	Nucleation Mode Aerosol Size Spectrometer (NMASS)
Large Aerosol Concentration	White Light Scattering (Climet & LTI/LasAir)
Sub-micron Aerosol Scattering & Backscattering (450, 550 and 700 nm)	TSI 3563 Nephelometer
Sub-micron Aerosol Absorption (450, 550, 700 nm) dry	Modified Particle Soot Absorption Photometer (PSAP)
Backscatter as f(RH) at 2 humidities	Radiance Research Nephelometers
Actinic Flux	Spectrally Resolved Radiometer, Zenith & Nadir
Broadband Radiation	Pyrgometer
Broadband Radiation	Pyranometer
Solar Spectral Irradiance	Solar Flux Radiometer
SO ₂ , O ₃ , H ₂ O column	Miniturized Differential Absorption Spectroscopy (MIDAS)

Table 4. Continued

Parameter	Method
Water Vapor (H ₂ O)	Tunable Diode Laser (TDL)
Water Vapor (H ₂ O)	Lyman Alpha Absorption
Air Temperature	Platinum Resistor
Dewpoint/Frostpoint	Dew-/Frost-point Hygrometer
Altitude	Barometric, Radar, GPS

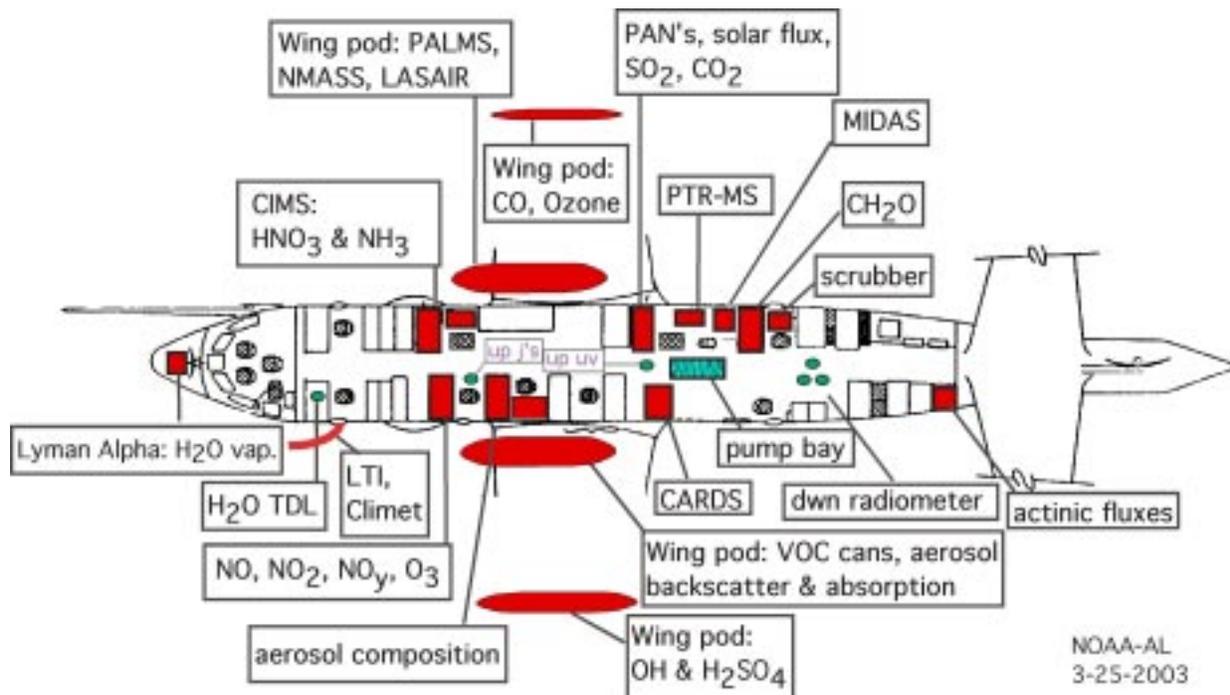


Figure 17: Proposed instrument layout for NOAA WP-3D Orion aircraft.

Acronyms:

CARDS	Cavity Ring-Down Spectroscopy (NO ₃ , N ₂ O ₅)
CIMS	Chemical Ionization Mass Spectrometer
LASAIR	Optical particle counter
LTI	Low Turbulence Inlet
MIDAS	Miniturized Differential Absorption Spectroscopy
NMASS	Nucleation Mode Aerosol Size Spectrometer
PALMS	Particle Analysis by Laser Mass Spectrometry
PTR-MS	Proton Transfer Reaction Mass Spectrometer
TDL	Tunable Diode Laser
VOC	Volatile Organic Compounds

Operations

AOC operates the WP-3D aircraft under VFR and IFR conditions. Since we are using external wing stores that don't have icing protection, flight into known icing conditions is not permitted. Minimum operating altitudes are determined by actual local flight conditions, but cannot override FAA regulations. Over water lower limits are determined by encountered conditions and location (obstacles like oil platforms, ships etc.). We have in the past sampled over water for short periods at 150 ft above sea level. Over land, flying missed approach patterns over local airfields lowered minimum operating height.

Usual Flight Planning schedule

Over the last 8 years we have used a WP-3D aircraft for air chemistry and aerosol research. From this experience an operational schedule for flight planning has evolved.

On the day before a planned flight:

1300 hrs local: After discussions with the modelers the flight planner submits the requested flight plan to AOC for review and for AOC coordination with the appropriate FAA and military authorities.

After submission this flight plan still can be fine-tuned to react to changes in weather and/or model forecasts.

Late in the afternoon: pre-flight flight readiness and flight goals briefing for all instrument PI's and project science team. At this time the flight planner reviews latest available weather and model forecast and adjusts the flight plan if necessary.

On the day of the flight:

3-4 hrs before take-off: The flight planner reviews latest weather and model forecast and adjusts flight plan if necessary.

About 2 hrs before take-off flight crew (pilots, navigator, and flight director) are briefed on final flight plan, last adjustments are discussed. After this meeting the flight crew files the flight plan with the FAA and confirms with military contacts if necessary.

Flight Plans

Preliminary flight plans have been developed. In any given flight, the focus will be on more than just one objective or science question. The flight plans will use the allotted resources (i.e. flight hours) in the most prudent way by addressing as many questions and/or objectives as possible on each flight.

The flights will reach into the suspected source regions to study the primary emissions of point and urban sources by gathering information on the signatures of the primary emission mix. This allows, by comparison with existing emissions inventories, to verify them. In the downwind regime the flights will follow the plumes advected to the North Atlantic and study their dispersion and the chemical and physical conversions within the plumes with progressing transport times.

Actual flight plans can only be finalized in the field according to the encountered meteorology and transport regimes. Nevertheless, some flight strategies & patterns have been developed over the years and they will be used in the NEAQS-ITCT 2004 study. Since several objectives/questions will be addressed in each flight, several strategies & patterns might be combined.

Figure 18 shows the aggregate flight tracks from the NARE 96 study. This study was conducted in March and April of 1996. It was also centered in the New England area and the figure shows the actual range of operation achieved. The NARE 96 study also investigated transport of pollution towards NE and its fate when advected to the North Atlantic, though especially the transport mechanisms of cold fronts.

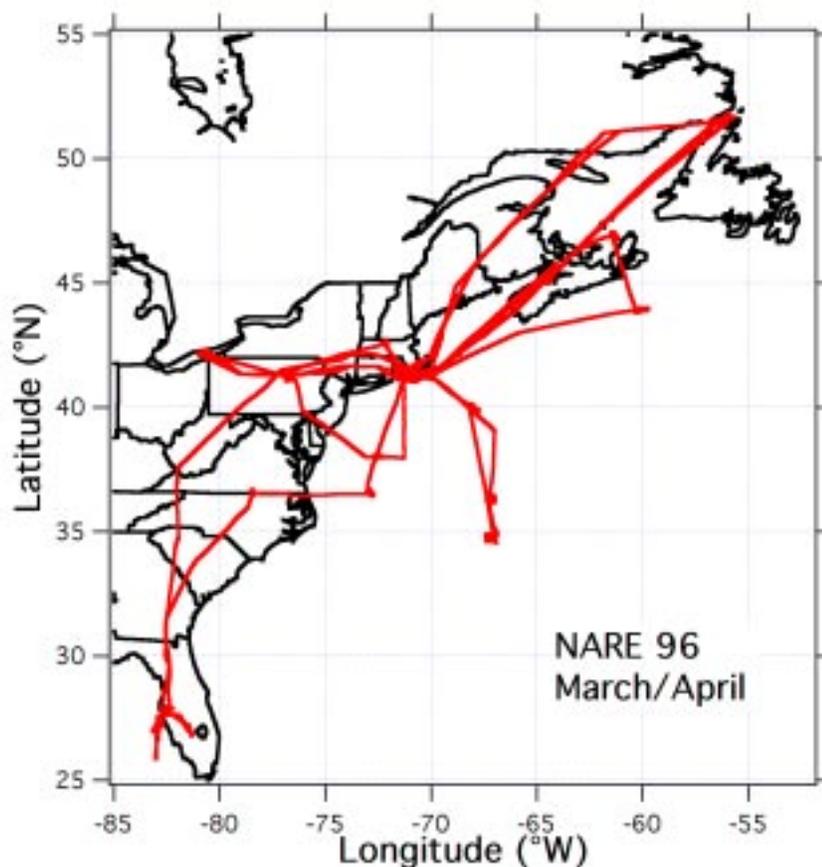


Figure 18: Flight Tracks from the NARE 96 Experiment

Some of the flight strategies & patterns that have been employed in the past are discussed here as an example to how they are providing data to NEAQS-ITCT 2004 study goals.

Flights upwind and downwind of power plants, and flights upwind and downwind of urban centers within the boundary layer and slightly above the boundary layer top to assess net fluxes from such sources and to identify the source signatures.

In a quasi-Lagrangian approach we have intercepted urban and point source plumes during downwind transport at various stages of their lifetime and recorded the photochemical and chemical conversions of the trace gases and the aerosol within those plumes, and investigated generation of aerosols within plumes.

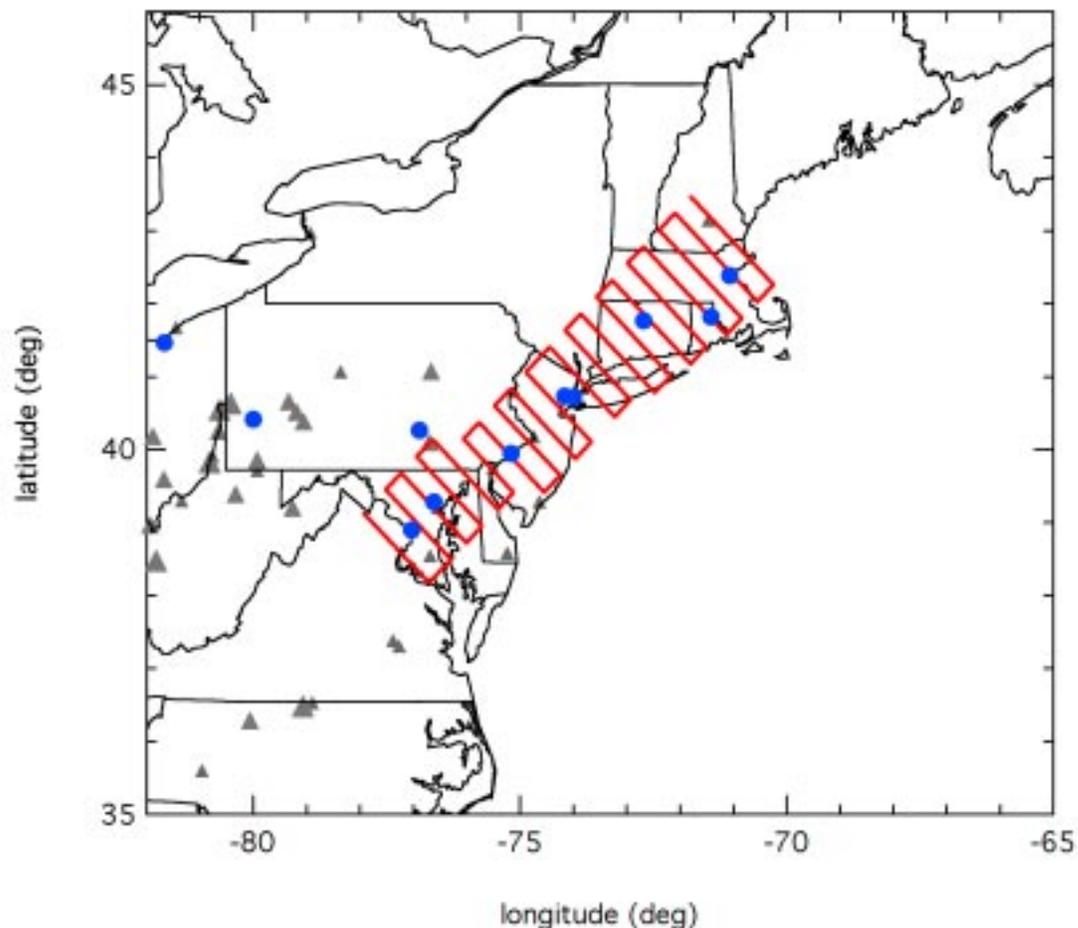


Figure 19. Potential flight track to study the evolution of emissions along the urban corridor from Washington, DC to Boston under flow from the southwest. At a wind speed of 5 m/s an air mass will travel from Washington to Boston in about 35 hrs. The flight track as shown is 3900 km, which is clearly too long for a single flight mission. Shown as blue circles are the locations of select urban areas. Shown as grey triangles are the locations of major NO_x point sources.

Flights within the boundary layer and above will address how much primary pollution is exported into the free troposphere. Around urban centers the impact of the heat island on boundary layer / free troposphere exchange will be investigated.

Flight patterns below, within, above and downwind of clouds will add information on cloud processing.

Flight patterns below, within, and above aerosol layers will add information on the impact of various kinds of aerosol on the radiation budget.

Flights above *Ronald H. Brown* will be used to validate remote sensing measurements made from that platform and answer how representative the ship measurements are for the entire marine boundary layer (MBL).

Coordinated flights below the flight track of the lidar aircraft will validate the lidar measurements and give an indication of how representative the *in-situ* WP-3D measurements are.

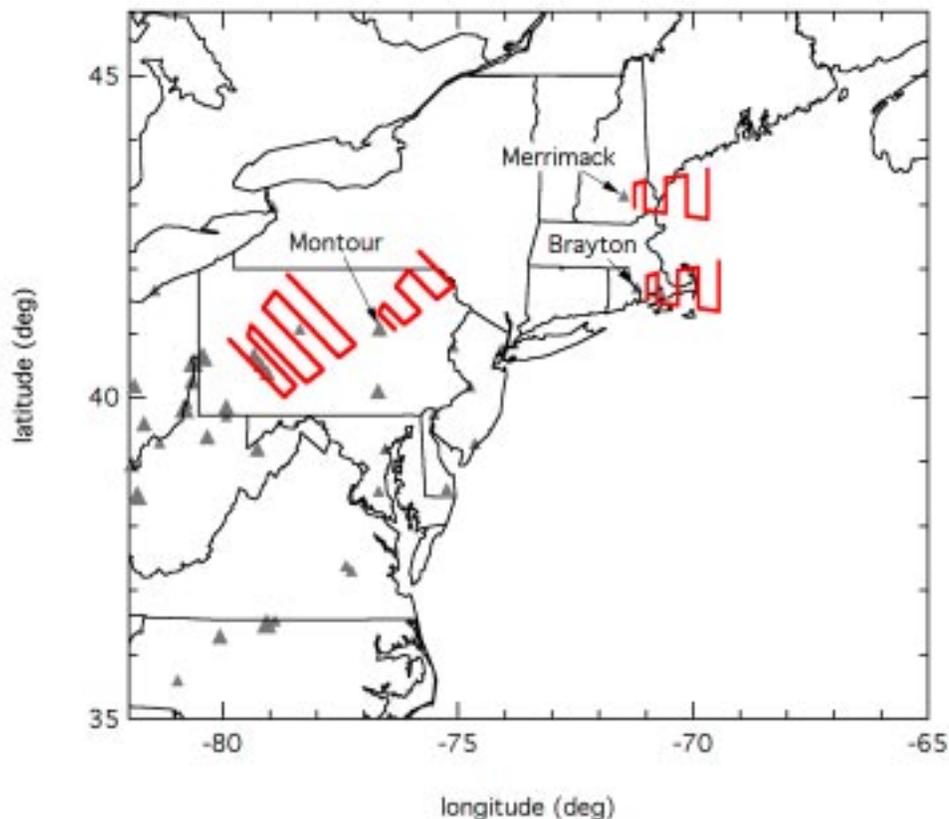


Figure 20. Potential flight tracks to study power plant plumes in New England (Merrimack, NH and Brayton, MA) and Pennsylvania (Montour, Homer City, Conemaugh, Keystone, PA) under a 5 m/s wind from the west and southwest respectively. The last transect shown is 144 km downwind from the power plants which for a nominal wind speed of 5 m/s corresponds to a plume age of 8 hours. Shown as grey triangles are the locations of major NO_x point sources.

Side by side flight at various altitudes in concert with other *in-situ* aircraft will intercompare the similar instruments on those platforms and assure that the generated data sets are compatible for the use by modelers.

Coordination with lidar aircraft

The capabilities on the *in-situ* (WP-3D) and the remote sensing (lidar) aircraft are complementing each other. Therefore, flights on any given day will be coordinated between the flight planners of the WP-3D and the lidar aircraft during the flight planning process in the field to maximize the synergies and take advantage of having these two valuable resources available simultaneously.

Remote Sensing Aircraft

In addition to the NOAA WP-3D aircraft, the 2004 experiment will feature a second aircraft focused on the application of differential absorption lidar (DIAL) techniques for remote sensing of local and regional ozone and aerosol. Previous field studies have benefited greatly from airborne measurements of ozone and aerosol profiles to characterize the 3-dimensional structure of pollution plumes and measure variability in mixing layer height (Senff et al, 1998, Banta et al, 1998). Airborne remote sensing enables tracking of plumes from urban areas and point sources, identification of isolated regions and layers of high ozone concentration, observations of atmospheric layering as characterized by aerosol structure, and investigation of local meteorological effects such as sea breezes and orographic lifting on pollution transport and mixing. For example, Figure 21 shows a time-height cross-section of ozone levels measured at the edge of Galveston Bay during the 2000 Texas Air Quality Study in the region around Houston, TX. The high concentrations and vertical transport of ozone due to convergence at the sea breeze front are clearly seen in the figure.

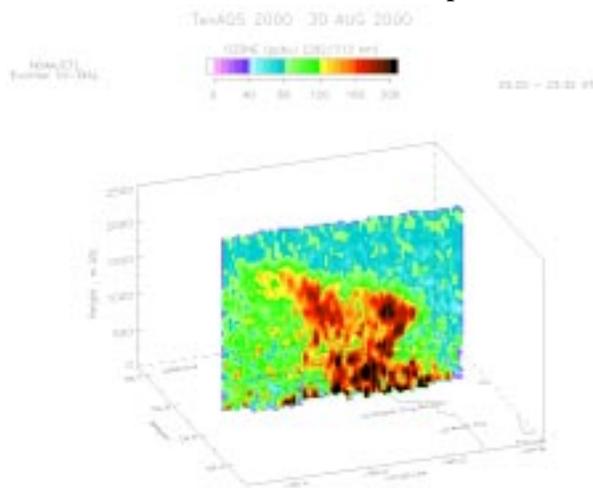


Figure 21: Time-height cross section showing high ozone concentration air transported vertically within the sea breeze front at the western edge of Galveston Bay.

Inclusion of a remote sensing aircraft also provides information on the 3-dimensional representativeness of *in situ* observations made on the WP-3D aircraft during those periods when the flight tracks of the two aircraft sample the same region.

Remote Sensing Aircraft Objectives

Flight plans for the remote sensing aircraft will specifically address scientific objectives associated with transport and evolution of pollution plumes, boundary layer structure, air quality forecasting and intercomparison of observations. Among the specific questions that the remote measurements will attempt to answer are:

1. *What is the spatial distribution of ozone and aerosol in the New England study region under different meteorological conditions?*
2. *What are the dimensions and structure of New York and Boston urban plumes as they advect across the study region?*
3. *How do plumes evolve as they travel over different land and water surfaces?*

4. *How does the sea breeze affect the structure and transport of ozone and aerosols at different locations on- and off-shore?*
5. *What (if any) vertical mixing takes place over the land and ocean during pollution events?*
6. *What is the effect of orography on the vertical transport of ozone and aerosol?*
7. *How do changes in land and water skin temperature affect the vertical structure of plumes advected over the ocean?*
8. *What is the impact of local urban sources, e.g., Portland, and isolated sources such as pulp mills on regional pollution distribution?*
9. *How representative are in situ measurements from ship and other aircraft platforms?*
10. *How well do air quality forecast models predict ozone levels, plume dimensions and transport, and atmospheric structure across the study region?*

On-board Sensors

The primary instrument to be deployed on the ETL-chartered aircraft will be the down-looking NOAA ozone/aerosol DIAL system (Alvarez et al, 1998, see Figure 22), which produces profiles of ozone and aerosol structure in the boundary layer and lower troposphere. The lidar system employs four wavelengths (277 nm, 292 nm, 313 nm, 319 nm) in the ultraviolet spectral region for ozone measurements. Each wavelength is characterized by a different ozone absorption cross-section, enabling measurements to be made over a wide range of ozone values. The multi-wavelength capability of the system also provides flexibility for correction of potential errors in ozone calculations caused by aerosol backscatter gradients. Ozone measurements are typically made at a horizontal resolution of approximately 600 m and vertical resolution of 90 m. Precision of the ozone measurement ranges from about 3 to 10 ppbV, depending on range and amount of intervening ozone. A fifth system wavelength at 360 nm, which is minimally absorbed by ozone, is used to measure aerosol backscatter profiles; resolution for the aerosol measurements is 600 m horizontally and 15 m vertically. Onboard the aircraft, a global positioning system provides a precise location for each lidar measurement, as shown for the plume tracking measurements in Figure 23. Data are analyzed and displayed on board the aircraft in real time, enabling adjustment or changes in the science mission if unexpected features or events are observed.



Figure 22: Airborne ozone and aerosol lidar deployed in NOAA-chartered de Havilland Caribou.

A key measurement objective for the New England study will be characterization of the structure of the boundary layer, including mixing layer height. Mixing layer height is estimated from the gradient of the lidar aerosol signal, as indicated in Figure 24. Investigation of mixed layer properties over different surfaces and the relationship with ozone concentrations will be important for understanding layering, transport, and vertical mixing. To provide additional information on surface properties, we also plan to mount a downward looking infrared radiometer to measure surface skin temperature on the aircraft alongside the ozone lidar.

At this time, we are investigating the feasibility and cost of incorporating a dropsonde capability on the ETL remote sensing aircraft. Dropsondes, particularly over the ocean, will provide information on wind structure associated with pollution layers, providing important information on the potential source of plumes observed distant from known sources. Because the aircraft will be flown unpressurized, installation of a dropsonde-launching configuration is anticipated to be relatively economical.

Aircraft Platform

The specific aircraft platform to be used for the lidar and other sensors will be determined in early 2004 after a formal procurement process. Past experiments have incorporated a CASA 212, a de Havilland Caribou (Figure 22), and a Douglas DC-3 to transport the lidar. The ETL remote sensing aircraft will tentatively be based at Pease International Tradeport and will fly at approximately 3-4 km altitude for missions extending over several hours. Primary requirements for the aircraft are:

- Capability to fly slowly (less than 75 m s^{-1})
- Capability to provide a nadir port for lidar installation
- Sufficient power and load capability for lidar and other sensors
- Over-water capability
- Mission duration greater than 6 hours

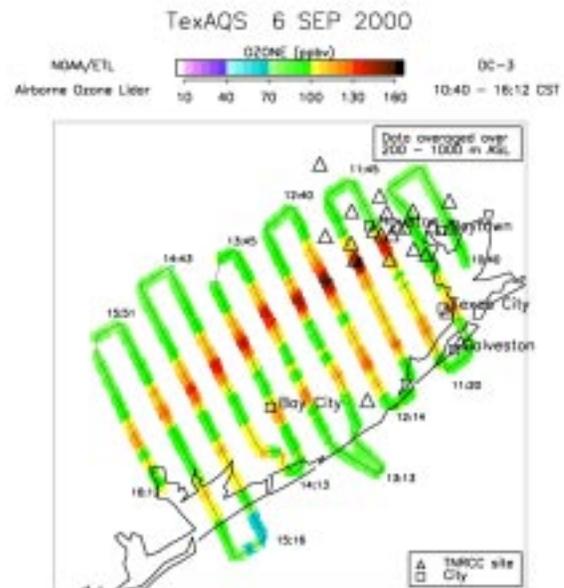


Figure 23: Flight track of lidar plume tracking measurements obtained during the Houston experiment. Ozone values are averaged over the layer between 200 and 1000 m AGL

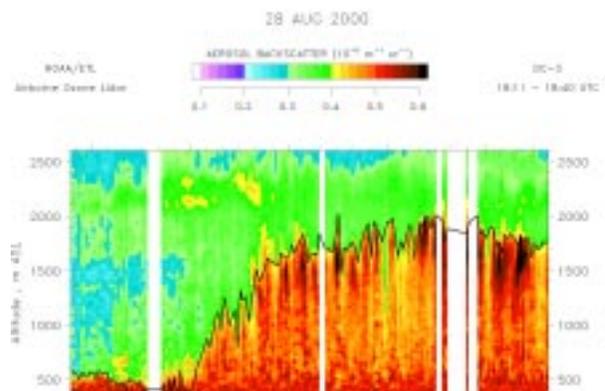


Figure 24: Vertical profile of lidar-observed aerosol backscatter showing a sharp change in mixing layer height at the Gulf of Mexico coast.

Aircraft operation and potential flight tracks

The aircraft will fly above the boundary layer at 3-4 km altitude to enable complete characterization of low level aerosol and ozone structure. Mission durations are expected to be on the order of 6 hours, depending on the aircraft selected for the experiment. As in previous experiments, daily flight tracks and mission objectives will be determined in collaboration with other experimenters on the day prior to the flight. Figure 25 (below) shows some candidate flight tracks for different science objectives.

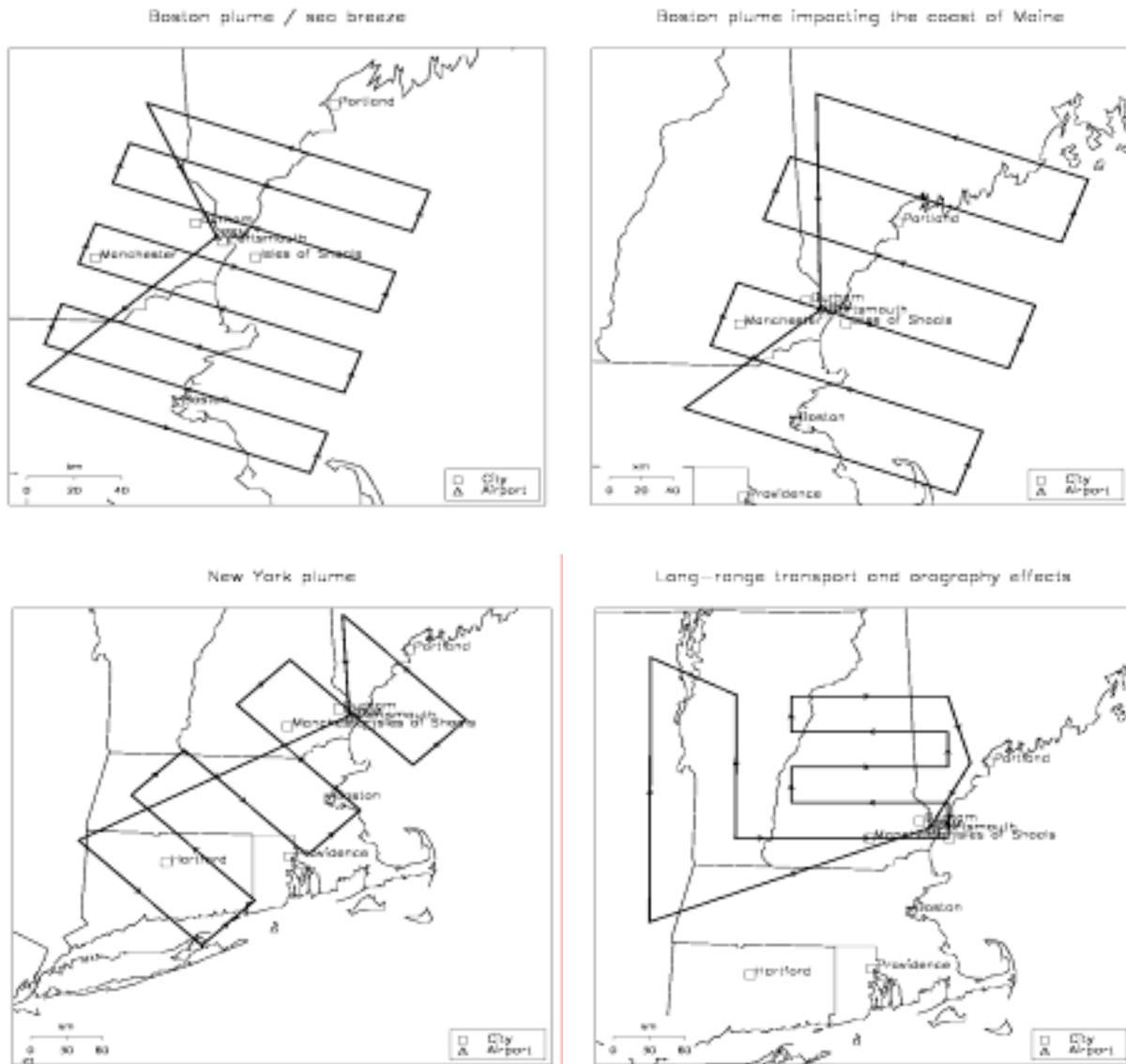


Figure 25: Proposed airborne lidar flight tracks for NEAQS 2004 Experiment

Profiler Network

Relevance

Transport in the coastal regions of New England is complicated by interaction between the continental and marine boundary layers and, in particular, the land-sea breeze circulation. Wind profiler and surface observations collected during NEAQS 2002 demonstrate part of this complexity. Figure 26 shows diurnally averaged wind profiles from Appledore Island, ME and Concord, NH for the months of July and August, 2002. The diurnal wind pattern at Appledore Island reflects a land-sea breeze circulation, with a synoptic southwesterly flow occurring at low levels during the daytime, on average, and synoptic northwesterly flow occurring during the night. The sea-breeze transition is indicated by a minimum in boundary-layer wind speed near midday (1500 UTC). The daytime sea-breeze flow at Appledore Island is favorable for transporting pollution from Boston and possibly other East coast metropolitan areas. Convergence caused by offshore flow from the continent and the sea-breeze flow may also contribute to increasing pollutant concentrations in the coastal zone. At Concord, the diurnally averaged wind directions reflect the mean synoptic forcing encountered during NEAQS 2002. The wind speed pattern shows the expected diurnal behavior over land in the summertime. Low-level speed shear occurs at night, a behavior caused by frictional decoupling of the surface from the winds aloft and stabilization of the nocturnal boundary layer. This shear is mostly eradicated during the day as solar heating drives mixing in the convective boundary layer.

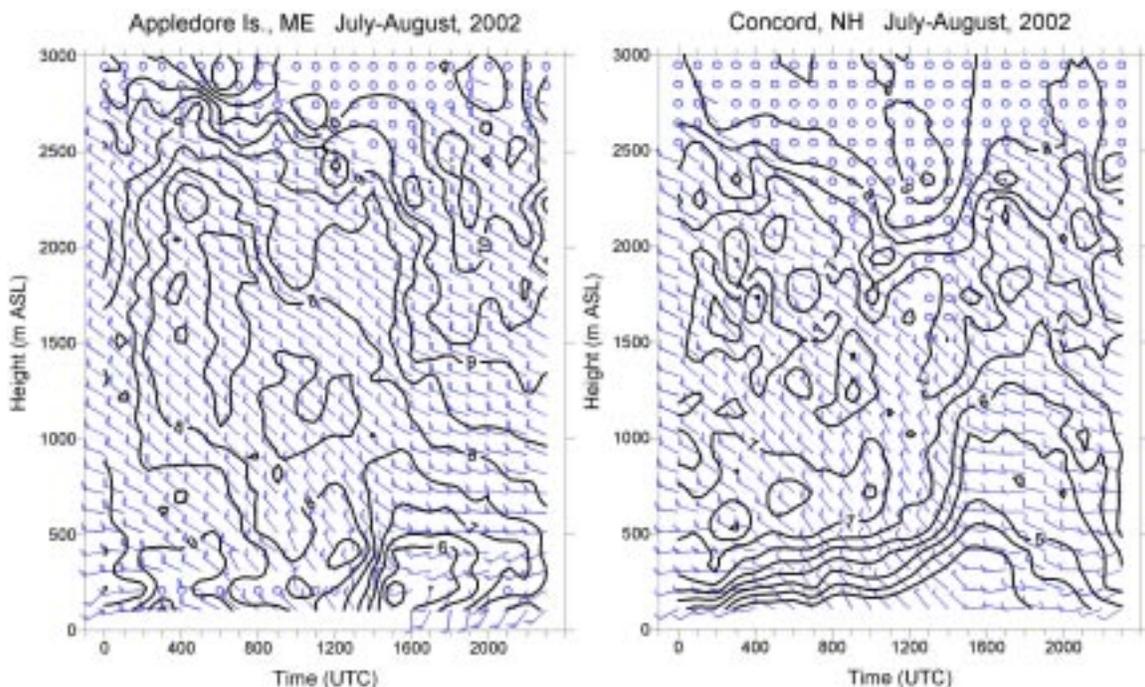


Figure 26. Diurnally-averaged wind profiles from the boundary-layer wind profilers deployed at Appledore Island, ME and Concord, NH during NEAQS 2002. Wind barbs show the vector-averaged wind direction. Contours (m s^{-1}) indicate scalar wind speed.

The contour interval is 0.5 m s^{-1} . Analysis includes all available data from July and August, 2002. Time-altitude points where data availability is less than 50% are indicated by open circles.

During NEAQS 2002 a vertically-pointing, unattended, ozone lidar was deployed alongside a wind profiler at Pinnacle State Park, NY. The main objectives of this deployment were a) to document the vertical structure of ozone and aerosol at a site upwind of the NEAQS 2002 study area and – in conjunction with the wind profiler data - to characterize the pollutant transport aloft from sources in the Midwest to the New England region, b) to assess the performance of the unattended ozone lidar over an extended period of time under field experiment conditions. As an example, figure 27 shows preliminary ozone data obtained with the unattended lidar at Pinnacles State Park on 24 July 2002. Generally, the altitude coverage for both ozone and aerosol measurements extended from about 200 m to 1800 m above the surface. The vertical resolution was 100 – 400 m (increasing with altitude) for the ozone measurements and 7.5 m for the aerosol measurements. The time resolution was 1 hour for both ozone and aerosol.

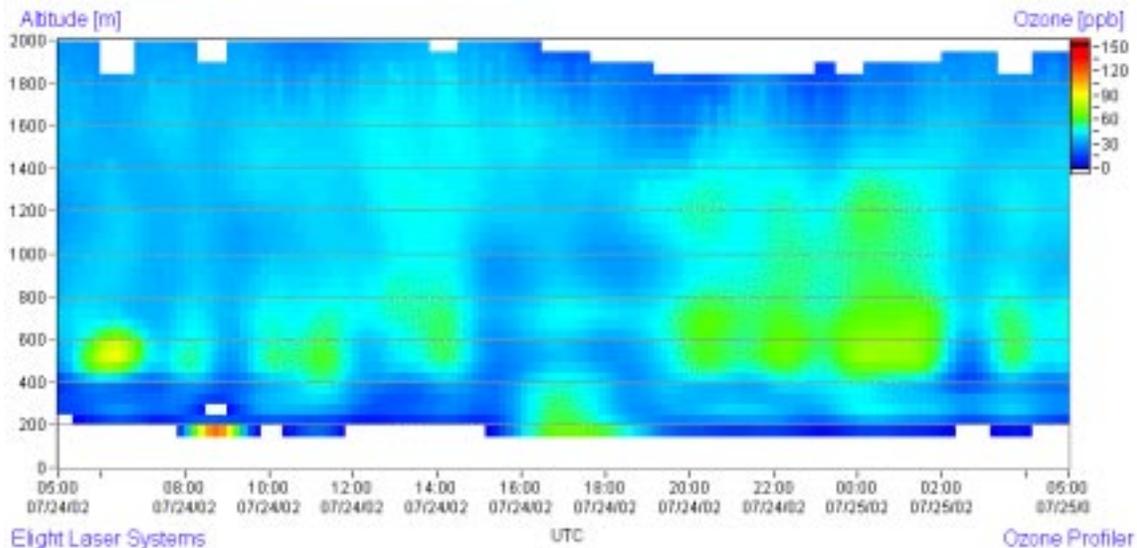


Figure 27. 24-hour time height cross section of ozone mixing ratio observed with the unattended ozone lidar at Pinnacles State Park on 24 July 2002.

Vertical mixing over the ocean is often inhibited by stable conditions near the ocean surface caused by warm continental air passing over cooler water. Well offshore, therefore, transport primarily occurs above the marine boundary layer. However, while *Ronald H. Brown* was close to the coast during NEAQS 2002, signatures of pollutant transport from the land often were observed at instrument level on the ship. The combination of wind profiler, ozone lidar, doppler lidar, and rawinsonde profilers

measured on the R/V Brown will be used to characterize mixing and transport over the ocean. In addition, shipboard measurements of the air-sea interface, including turbulent fluxes of heat, momentum, radiation, and possibly ozone, will enable estimates of deposition over the ocean. As in NEAQS 2002, ship tracks near the coastline will help reveal the interaction between continental and marine boundary layers.

During NEAQS 2002, surface energy budget observations were conducted at the Thompson Farm chemical sampling site in Durham, NH. The radiation data collected from this site have revealed an important result regarding the relation between aerosols and solar radiation and the importance of the treatment of aerosols, or lack thereof, in numerical models used for weather prediction. Figure 28 compares modeled and observed solar irradiance as a function of aerosol optical depth at 1400 UTC on 5 days during an ozone episode from NEAQS 2002. The lack of aerosol treatment in the ETA model leads to a $\sim 84 \text{ W m}^{-2}$ bias between observed and modeled solar irradiance. Incidentally, using a simple radiation balance model, an 80 W m^{-2} error leads to a 2° C

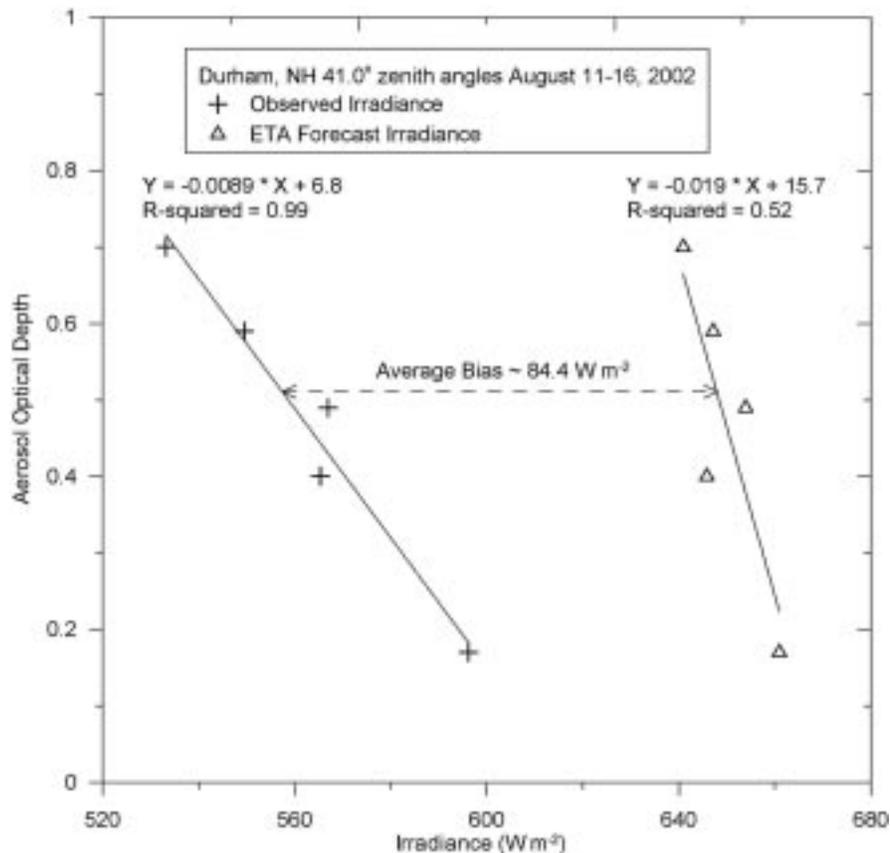


Figure 28. Observed versus modeled solar irradiance as a function of aerosol optical depth at 1400 UTC during the mid-August 2002 ozone episode. The measurements of solar irradiance and aerosol optical depth are from the Thompson Farm air chemistry site in Durham, NH. The ETA model does not include the effect of aerosols in computing solar irradiance. The bias between observed and modeled irradiance corresponds to $> 2^\circ \text{ C}$ error in skin temperature based on a simple radiative balance.

error in skin temperature. An unknown aspect of these results is the contribution from the water vapor path. For example, the reason that the ETA solar irradiance shows any positive correlation to aerosol optical depth may be due to the impact of the water vapor in the atmosphere. For the 2004 study, a complimentary set of radiometric sensors is planned for *Ronald H. Brown* and at Concord, NH, chosen to be far enough inland to avoid any influence of marine aerosols. This research addresses one of the NEAQS and ITCT goals of investigating the direct radiative effects of aerosols in the atmosphere, as discussed in the Chemical Transformation section.

Science Questions

The integrated boundary-layer wind profiler observing system network for NEAQS-ITCT 2004 is designed to address the following key science questions:

1. *What are the main transport corridors that bring transported air pollution into the Northeastern U.S.?*
2. *How are the continental and marine boundary layers linked and how does this coupling affect the vertical distribution of pollution transported over the ocean?*
3. *Where and when is the sea-breeze/land-breeze circulation important?*

In addition, data from the profiler network will be used in conjunction with model output to evaluate the skill of model forecasts and to uncover deficiencies in the models and quantify their impact. Key questions with respect to numerical models are:

4. *How well do operational and research models used for weather and air quality prediction reproduce the observed meteorology (diagnostic and operational evaluation)?*
5. *What improvements in model design and/or operation are needed to significantly improve forecast skill?*

Deployment Strategy

The design of the integrated wind profiler observing system network is driven by two primary and somewhat competing objectives outlined in the Transport and Mixing section: to characterize in 2004 and to develop a climatology of boundary-layer structure along the New England coast (i.e., an ABL-scale issue), and to measure lower tropospheric winds with sufficient horizontal resolution to characterize transport corridors in the Northeast (i.e., a regional-scale issue). The first objective is important to understand the vertical distribution of pollutants, especially in the coastal zone, and to place the relatively short field programs of 2002 and 2004 into a climatological context. The second objective addresses one of the primary science questions for NEAQS-ITCT 2004: where does air pollution measured in the Northeast originate? These objectives should be met by the proposed regional network of approximately fourteen wind

profilers. Part of this network will be provided by cooperative agencies that run profilers in the region, as was the case in the NEAQS 2002 study. In particular the profilers at Stow, MA, New Brunswick, NJ, Fort Meade, MD, Richmond, VA, and Lunenburg Bay, Nova Scotia have been included in network plans for 2004. Unfortunately prior experience, including experience gained in NEAQS 2002, has shown that many of the state agencies that have purchased wind profilers do not have sufficient funds or the expertise to properly site, operate, and maintain their equipment. Close coordination between the NEAQS Science Team and the other agency profiler operators prior to and during the 2004 field study will be necessary to ensure that reliable profiler datasets are collected from cooperative agency sites. In addition, the 2004 study will take advantage of other meteorological measurements collected by non-NOAA public and private entities. The Ground Winds Lidar operated at Bartlett, NH by the University of NH is an example of an instrument that could provide detailed meteorological information to enhance the scientific research planned.

In addition, we are planning to deploy collocated with wind profilers two vertically pointing unattended ozone/aerosol lidars contingent upon the availability of suitable instruments. The lidars would provide important information on the vertical structure of ozone and aerosols and pollutant transport aloft thus contributing to one of the key objectives of the 2004 study of identifying and quantifying pollution transport pathways into the New England area. We envision to place one of the unattended lidars at an upstream site west of the main study area to characterize the influx of pollutants from the Midwest. We plan to deploy the second lidar in the vicinity of the New England coast, where the lidar and collocated wind profiler could provide important information on the impact of smaller-scale circulations, such as the land/sea breeze flow, on pollutant distribution in the New England area.

Figure 29 shows the planned locations of integrated boundary-layer wind profiler observing systems and vertically-pointing unattended lidars that will be deployed for 2004 study. Five of the nine NOAA land-based wind profiler sites, including the three (Appledore Is., Pease, and Concord) that form a line perpendicular to the New Hampshire coastline, were operated during NEAQS 2002 and their operation possibly will be extended through the summer of 2003 (funding pending). New wind profiler sites selected for the 2004 study consist of Southern Ohio; Pittsburgh, PA; Bar Harbor, ME; and Sable Island, Nova Scotia.

Two new web-based tools will be developed for the study to assist scientists and forecasters in accessing and using wind profiler data. The first of these will be a menu driven profiler trajectory tool. This interactive tool will allow users to plot wind profiler-deduced trajectories at levels within and above the boundary layer. The users will be allowed to choose a location on a base map for either forward or backward trajectories and specify either current or archived data. The second tool will be a real-time display of profiler deduced boundary-layer depth. The algorithm used to create this display is based on a fuzzy logic technique that substantially improves wind profiler boundary-layer height detection (Bianco and Wilczak, 2002). A publicly available NOAA web page will

continue to provide access to integrated profiler observing system products from the study in near real time. This web page also will include a tool for evaluating the performance of forecast models by allowing users to compare forecasted profiles and surface values of relevant meteorological variables from an array of operational and research forecast models with observed quantities at model grid points corresponding to the locations of the integrated profiler observing system network.

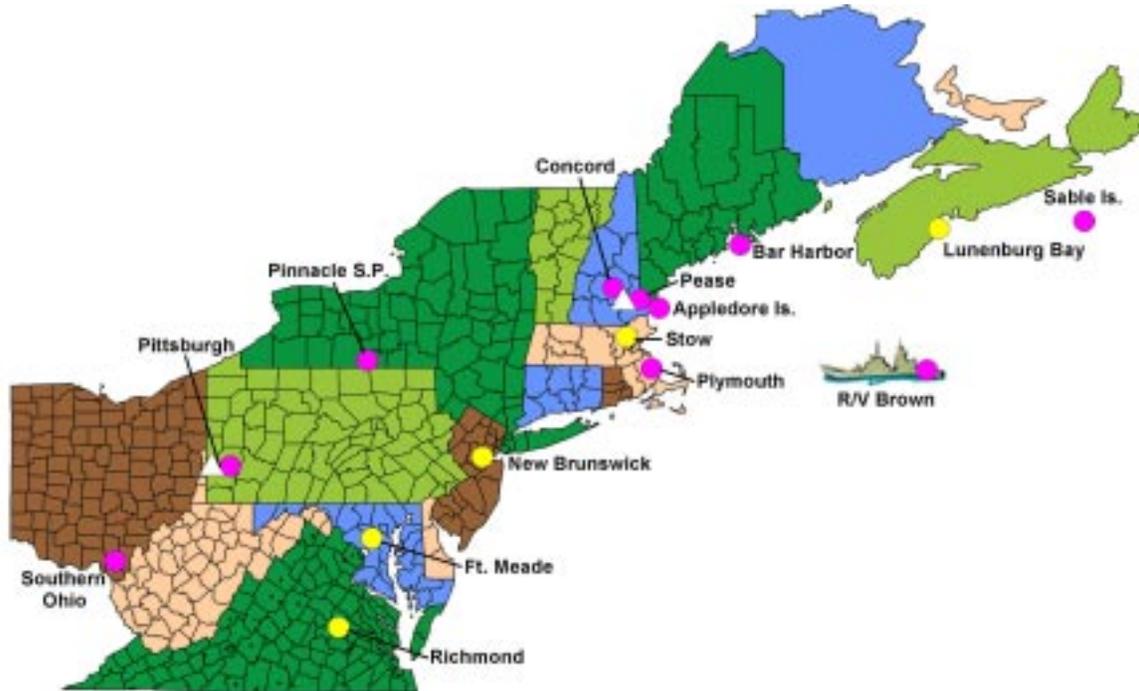


Figure 29. Proposed NEAQS-ITCT 2004 profiler network. Pink circles denote NOAA wind profilers, yellow circles denote cooperative agency wind profilers, and white triangles denote vertically-pointing unattended lidars. The full set of instrumentation on *Ronald H. Brown* is listed in a previous section.

Intercomparison Activities

Relevance

Quantifying measurement uncertainty establishes an objective, defensible basis upon which subsequent scientific interpretation can be founded. Intercomparison of similar measurements between the different NOAA platforms during the 2004 field program provides an opportunity to test and evaluate instrument performance to potentially identify any unanticipated operational problems, thereby tempering and improving interpretation of the resulting data sets.

Science questions

Intercomparison exercises are planned to provide answers to two basic questions:

1. *How well do the instruments actually perform in the 2004 mission?*
2. *Are different analytical approaches to a single answer mutually consistent?*

These questions are explored below.

1. Intercomparison involving well-established techniques or relatively mature instruments. This kind of field comparison is designed to test a particular *implementation* of a proven technique or techniques, rather than the techniques themselves. A quantitative comparison requires knowledge of the estimated uncertainty in the reported data, which will be provided prior to the exercise by the instrument operators. These uncertainties are typically derived from *internal* consistency checks, laboratory tests, and historical performance of the instrument in prior field missions. The intercomparison exercise provides an additional *external* consistency check of these uncertainty estimates; under an ideal situation, the data will agree to within the combined uncertainty of the measurements being compared. Discrepancies that lie outside of the combined uncertainties suggest possible unidentified sources of error in one, or both, instruments. In this case, analysis of ancillary data can sometimes give an indication of which, if either, instrument is functioning properly. (To help quantify estimated uncertainties prior to the comparison, operators will be asked to fill out a standardized information sheet describing relevant instrumental details).

2. Intercomparison involving emerging techniques or newly-developed instruments This exercise involves more recently developed instruments or those without a proven history in the field. Often these comparisons provide useful information on how a new technique can be improved for the future, as the investigators learn about how a given instrument actually behaves under the demanding conditions in the field. The estimated instrumental uncertainties may be less well known prior to the intercomparison exercise than those of the more mature, well-developed instrumentation, described above. The intercomparison of emerging techniques nonetheless still can provide an external check on instrument

performance for this particular mission, especially if a benchmark measurement from a well-established technique is available.

3. Intercomparison of derived values Occasionally, with redundant information available, there are consistency checks involving more than two instruments at once. An example of this is the comparison of the sum of individually-measured constituent NO_y species to the NO_y measurement itself. Another might involve a comparison of the aerosol optical depth measured from *Ronald H. Brown* to that calculated from *in-situ* measurements during an aircraft altitude profile above the ship. These so-called closure experiments can provide rigorous tests of our understanding of measurement uncertainties. They further provide realistic bounds on our ability to interpret the chemical and physical state of the atmosphere from ambient measurements.

Deployment strategy

Intercomparison exercises can take place before, during, and after the 2004 field mission; those most likely to provide useful and relevant information, given logistical and timing constraints, are briefly described below.

Evaluation of standards (pre, during, post-mission): Comparison of compressed gas standards should be performed at least once for the different *in-situ* gas phase instruments on the WP-3D and *Ronald H. Brown* for NO, CO, CO_2 , and SO_2 . Comparison of ozone standards, requiring circulation of a NIST-traceable ozone source, should also be performed. Effort will also be made to evaluate instrumental sensitivities to HNO_3 between the aircraft and shipborne instrumentation by sampling from a characterized permeation device. PILS IC standards will be exchanged between investigators on the WP-3D and *Ronald H. Brown*.

Direct comparison of measurements (pre, during, post-mission) Running instruments in the lab or in the field side-by-side is an excellent way to test performance. This is more easily done for some instruments than others, so this will be up to the various investigators to arrange as desired. During the mission joint flights of the two aircraft, and overflights of the ship, will provide data from which instrument performance may be critically assessed. The WP-3D will have duplicate measurements of O_3 (UV absorption and chemiluminescence), selected hydrocarbons (whole-air samples and PTR-MS), aerosol chemical composition (PILS and AMS), and possibly N_2O_5 (cavity ring-down spectroscopy and CIMS) that will be compared when possible on every flight. Joint WP-3D and ETL profiling lidar aircraft flights will permit comparison of *in-situ* ozone with the altitude-resolved ozone retrieval. Aerosol backscatter data may also be comparable between these two aircraft if the effects on backscatter of humidity changes during sampling can be taken into account. Overflights of *Ronald H. Brown* provide an opportunity to compare a large variety of gas-phase, aerosol-phase, meteorological, and radiative parameters. Comparison of *in-situ* WP-3D and ship data depends upon the

minimum altitude of the aircraft (~50 to 100m), which should be sufficient to sample all but the most shallow of marine surface layers.

Indirect comparison of measurements (during and post-mission) While sampling in close physical proximity provides useful information, other opportunities exist to evaluate instrument performance by examining data taken during normal flight procedures. For example, the CO/CO₂ ratio should be approximately conserved for some time during transport over water, suggesting that WP-3D and ship data in an urban plume might be usefully compared in this regard. Further, free tropospheric ozone levels should be comparable between the ETL aircraft and the WP-3D if taken within a relatively short volume and time of one another. These intercomparisons-of-opportunity can provide useful additional data with which to evaluate instrumental performance between the various NOAA platforms.

These NOAA-intramural intercomparisons will be informal but will be performed blind, to provide a real-world test of the instrument performance under normal field conditions. Each data set will be reduced and submitted independently, accompanied by the estimated uncertainties, before accessing the comparison data. The results of the various intercomparisons will be disseminated to all the study participants via presentations in the field science meetings and, following the mission, at the data workshop. Finally, a mention of the intercomparison and a summary of the results should be included in any publications that make use of the resulting instrumental data.

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