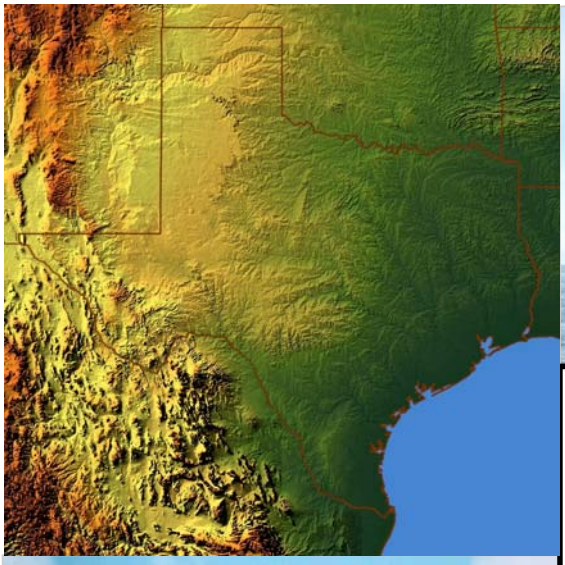
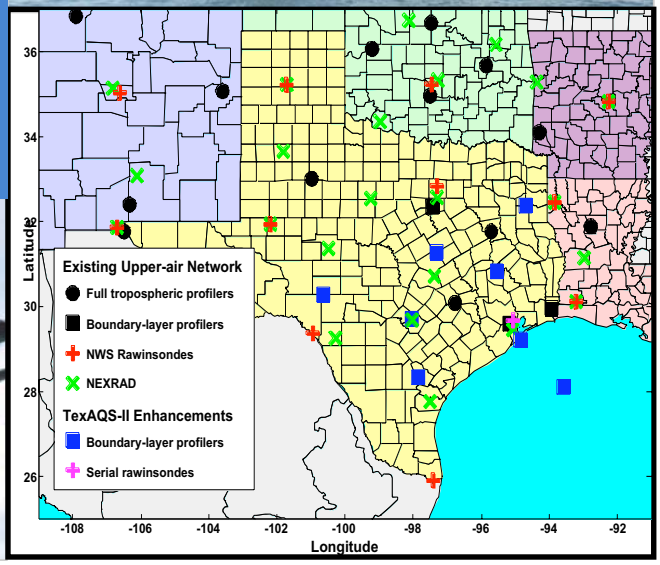
2006 TEXAQS/GoMACCS

Science and Implementation Plan





Combining Climate Change and Air Quality Research

March 2005

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Introduction

Meeting Society's Needs: The Aim of the 2006 Study

The challenge of properly managing our atmospheric resources is complex. Improved understanding to inform policy development and implementation is needed in several areas. NOAA has established the Climate and Air Quality programs to address these issues. These comprehensive programs have as one of their foci an improved understanding of the workings of the chemical processes in the atmosphere. Major components in both the Climate and Air Quality programs include laboratory studies, field measurements, and modeling studies. The field studies include long term monitoring, short term limited deployments and intensive field campaigns using multiple platforms and a large array of instruments.

As a part of this overall program in 2006, NOAA will help lead a major multi-institutional intensive field program that will focus on investigating important scientific questions that are common to both climate and air quality. The NOAA components of the program are the Texas Air Quality Study (TEXAQS) and the Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS). This intensive field study will focus on providing a better understanding of the sources and atmospheric processes responsible for the formation and distribution of ozone and aerosols in the atmosphere and the influence that these species have on the radiative forcing of climate regionally and globally, as well as, their impact on human health and regional haze. The study area will be Texas and the northwestern Gulf of Mexico.

The NOAA Climate Change Focus in 2006 – GoMACCS, the NOAA climate change component of this field program, will characterize marine/continental chemical and meteorological processes over Texas and the Gulf of Mexico in order to improve the simulation of the radiative forcing of climate change by lower-atmosphere ozone and aerosols. In addition to clear-sky radiative effects, GoMACCS will investigate the influence of aerosols on cloud properties and the role of clouds in chemical transformation.

The NOAA Air Quality Focus in 2006 –TexAQS 2006, the NOAA air quality component of this field experiment, will investigate the sources and processes that are responsible for photochemical pollution and regional haze during the summertime in Texas. Figure 1 indicates the counties in Texas that are experiencing air quality problems associated with this ozone. In addition, there is growing concern that additional counties in the state may be facing similar issues in the near future. The 2006 study will provide information on the sources of the ozone and aerosols precursors and processes responsible for the formation and distribution of ozone and aerosols in the state.

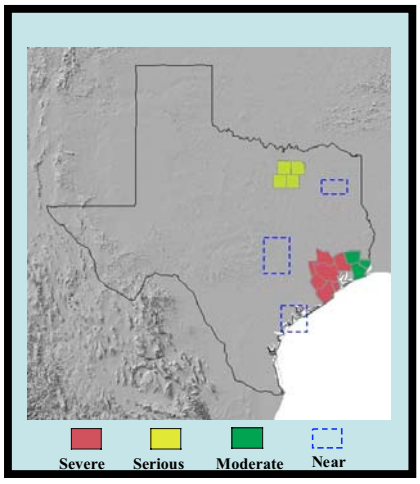

Fig. 1: Counties in Texas with air quality problems.

The focus of the study will be the transport of ozone and ozone precursors within the state and the impact of the long-range transport of ozone or its precursors into (and out of?) the state. In this regard, special attention will be paid to nighttime chemistry and transport. The study will also investigate how the various urban, industrial and natural sources of aerosols and aerosol precursors within the state and the transport of aerosols from outside the state contribute to the regional haze that is observed in the state.

The Importance of Ozone and Aerosols in Air Quality and Climate Change

Ozone and climate change - Ozone is an important trace gas in the troposphere and plays several important roles in determining the radiative forcing of the atmosphere. Ozone is (i) a radiatively important trace species (c.f., Fig. 2), (ii) important in climate feedback mechanisms, and (iii) important in controlling atmospheric oxidation. It is currently believed that a significant fraction of the ozone in the troposphere is produced by photochemistry within the region from precursors that have both human-influenced and natural sources. The lifetime of the ozone in the free troposphere is sufficiently long that it can be transported into the upper troposphere and over hemispheric scales. Thus, a predictive understanding of ozone and its influence on climate, as well as the influence of

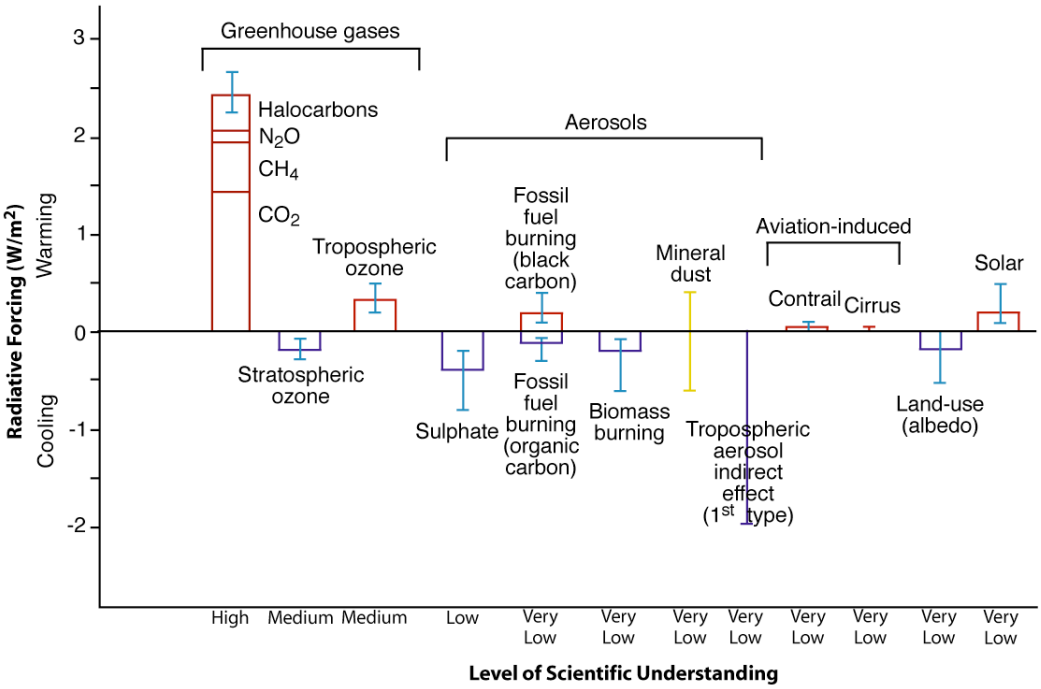

Fig. 2. Contribution to global warming/cooling for various atmospheric constituents (IPCC, 2001).

climate change on ozone and aerosols, requires a focus on the chemical and transport processes that link regional emissions to hemispheric ozone trends and distributions.

Aerosols and climate change – Aerosols force changes in the climate system not only via their absorption and scattering of radiation (the so-called "*direct effect*") but also through

indirect effects by changing cloud properties (c.f., Fig. 2). Namely, increasing the number of cloud-forming aerosols in the atmosphere, at a given amount of condensable water, tends to yield larger number of smaller droplets and brighter clouds (the so-called "*Twomey effect*"). Brighter clouds reflect more radiation back to space and hence tend to cool the climate system. Aerosols can also alter the precipitating capability and, hence, the longevity (and brightness) of clouds and atmospheric convection. The indirect effects of aerosols are currently deemed to be the most uncertain of all the forcings (See Figure 2). Research is needed to reduce the aerosol-cloud-climate uncertainty and determine the sources and evolution of aerosols in the atmosphere.

Ozone and air quality – Ozone plays a central role in determining air quality. From the standpoint of health effects, ozone is the principal pollutant associated with photochemical smog. Elevated and potentially harmful levels of ozone are observed in many areas of the United States during the summer. Although the highest levels of ozone are recorded in urban areas, and these are sufficient to be a human health hazard, ozone in rural areas can also do significant damage to sensitive vegetation. Of all the pollutants that are addressed in the Clean Air Act, ozone has proven to be the most difficult to control.

Aerosols and air quality – Studies have indicated that tens of thousands of people die each year in the United States as a direct result of exposure to high levels of aerosols, with many more suffering adverse health impacts that result in lost work and hospital visits. The aerosols found in polluted air can cause regional haze that obscures scenic vistas and present a hazard to aircraft(c.f. Fig. 3). As air pollutants fall back to earth they can acidify our lakes and streams and foul our coastal waters with serious consequences for these sensitive ecosystems (Lovett, and Lindberg, 1996; Fenn et al, 2003).

Fig. 3. A polluted haze covers downtown Houston.

The NOAA Approach: Combining Climate Change and Air Quality Intensive Field Campaigns

Until recently NOAA's research-intensive field campaigns in the Climate Forcing component of the Climate Program and the regional air quality component of the Weather and Water program have been conducted as separate, albeit related, activities. One focus of this research has been a better understanding of global-scale transport and transformation processes, which is linked to other national 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 aerosols in a number of settings including: Nashville, TN (1994, 1995, 1999), Atlanta, GA (1999), Houston TX (2000) and New England (2002, 2004).

The Texas study in 2000 is particularly germane to the 2006 Study. Scientific understanding gained from Texas 2000 Air Quality Study was of keen interest to decision makers since the region experiences some of the most severe episodes of poor air quality in the Nation. New insights emerged from the field study that relate to hydrocarbon emissions from the petrochemical facilities. Emissions of the hydrocarbons are estimated by industry using modeling approaches, and the Texas 2000 Air Quality Study data were of particular interest because they provided the opportunity to cross-check those emissions inventory estimates. Additional findings that elucidated the role of various and relatively unique sources of ozone and aerosol precursors in the region (such as transportation, the petrochemical industries, and other industrial sources), as well as the specific atmospheric processes associated with the region (in which departing air is "re-circulated" over the Gulf of Mexico and returned back to the land area) were analyzed and assessed.

Fig. 3. Major petrochemical industries are located in Houston.

During each of the previous intensives NOAA had specific climate or air quality goals. In these intensives NOAA operated instrumented aircraft and participated in specialized ground based and remote sensing measurements that were directed to achieving those specific goals. However, the distinction between the research objectives of the climate or air quality intensive field campaigns was, at least in part, simply a matter of perspective and scale. Many of the chemical and meteorological processes of interest were common to both. In addition, 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 conducted its first joint regional air quality and climate change study in the summer of 2004. The study combined the elements of the previous ITCT and NEAQS studies. The study focused 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) were being deployed in a manner that supported the objectives of both research programs. The TexAQS/GoMACCS intensive in 2006 is a continuation of this approach.

Research Areas

The research planned for the TexAQS/GoMACCS 2006 field campaign has been organized around the following five research areas, each with an associated science question.

Emissions verification and assessment - 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 Texas 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 aerosol in this region 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.

Science Objectives and Associated Science Questions

Emissions Verification and Assessment

Relevance

Improving the quantitative understanding of the location, timing, and speciation of gaseous 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 [*Jang and Fast, 2004*] and climate change processes [*IPCC, 2001*]. Reports suggest that while some U.S. inventories are reasonably accurate (e.g., point source NO_x and SO_2: [*Ryerson et al.*, 1998]), others may have substantial errors (e.g., on-road emissions of CO: [*Parrish et al.*, 2002]; petrochemical alkenes: [*Wert et al.*, 2003]). A goal of the NOAA 2006 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 petrochemical industrial facilities, on-road vehicles and from large electric utility power plants to anthropogenic gas- and aerosol-phase pollutants in the summertime Texas airshed are expected. Other potentially significant contributions may come from off-road transport such as commercial shipping. Substantial biogenic contributions to reactive VOC compounds involved in ozone and secondary organic aerosol formation are also expected.

The summer 2006 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., light absorbing carbon (LAC)) as well as gas-phase (e.g., VOCs, SO_2, CO, etc.) compounds. The top-down approach that we use provides an independent assessment of existing inventories. This approach has four distinct features:

> *Measurement of <u>ratios</u> of co-emitted pollutants compared with predictions derived from existing emissions inventories:* Many sources emit more than one species at a time. Measurements of the ratios of these species in the emission plume provides a check on the existing emission inventories, and may be used to identify a particular source among several possible emitters. Also, 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 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.

Measurement of absolute amounts of emitted pollutants compared with estimates derived from 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.

Measurements of spatial patterns of emissions compared with estimates derived from inventories: As an example, the geographic distribution of SO_2, NH_3, 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 light absorbing carbon (LAC) sources 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.

Measurement of temporal variations in emissions strength and composition compared with estimates derived from 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, including diurnal cycles and weekday-weekend differences. Variability in either can influence the chemical composition and radiative properties that result from their release to 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.

Based on this top down approach, we hope to address the following five specific science questions during the 2006 intensive.

1. What is the regional distribution of anthropogenic and biogenic emissions in Texas?

The sources of the precursors of ozone and aerosols are widely distributed in Texas. This is illustrated in Figure 4 which shows the distributed anthopogenic CO emissions and major point sources of NO_x in Texas. In the figure, area wide CO emissions are color-coded according to the intensity of the sources and the point sources are indicated as red circles with the magnitude of the source proportional to the size of the circle (c.f., Emissions Web Browser [http://map.ngdc.noaa.gov/website/al/emissions/], NOAA,

National Geophysical Data Center). The spatial distribution of the CO emissions, are concentrated in the large metropolitan Areas of Dallas - Fort Worth, Houston and San Antonio - Austin. These metropolitan areas account approximately 70 % of the CO emissions over Eastern Texas. The rest of the CO emissions come from distributed emissions along the highways connecting these cities and from smaller urban centers as well as a few significant point sources.

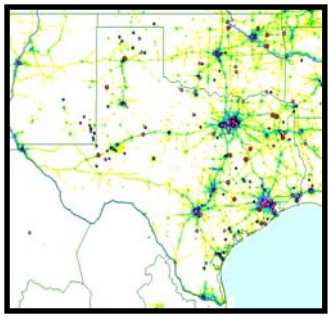

Fig. 4. The spatial distribution of CO and NOx emissions in Texas.

In addition to the area CO emissions, the figure also shows the location of large industrial NOx point sources *(indicated by the red circles in the figure),* such as electric power generating units. Several of the large power plants in east Texas are located in the forested, rural areas of northeastern Texas and between Dallas and Houston. Hence, the potential for ozone formation on a regional basis will rely on the distribution of the reactive biogenic VOCs. According to the EPA National Emission Inventory (NEI) 1999 these point sources contributed approximately 45 % of the NOx emissions in Eastern Texas. However, since the 2000 study the NOx emissions from many of the power plants have been substantially reduced through the installation of control equipment.

The implication of the distribution of these sources, their relation to each other and the changes in emission with time play an important role in assessing their contribution of the regional distribution and long-range redistribution of ozone and aerosols. The use of top-down measurements can help the assess the current situation and changes that have occurred.

2. What are the relative contributions from urban area sources, power plants, and industrial emissions within the Houston metropolitan air shed?

What sets Houston apart from other urban areas of its size, like for example Dallas – Fort Worth and Atlanta, is the presence of the petrochemical complex along the Ship Channel and Galveston Bay with its collocated emissions of NOx, SO_2, and reactive hydrocarbons. The impact of these collocated emissions on the measured mixing ratios of several gas-phase species is illustrated in Figure 5. On 28 August 2000 the winds during the morning hours were from the south-southeast at about 4-5 m/s and the measurement across the Houston airshed along the transect at 29.8 N give a good representation of the contribution of the major emission sources. On the western side of the transect, the plume of the coal and gas fired W.A Parish power plant with a generating capacity of 3600 MW can be clearly seen in the traces of the NOy, SO_2 and CO_2 concentrations. On the eastern side of the transect, the gas and oil fired Cedar Bayou power plant with a generating capacity of 2260 MW leads to a plume of enhanced NOy and CO_2 concentrations, but low SO_2 levels. While the urban center contributes noticeably to the NO_x emissions in

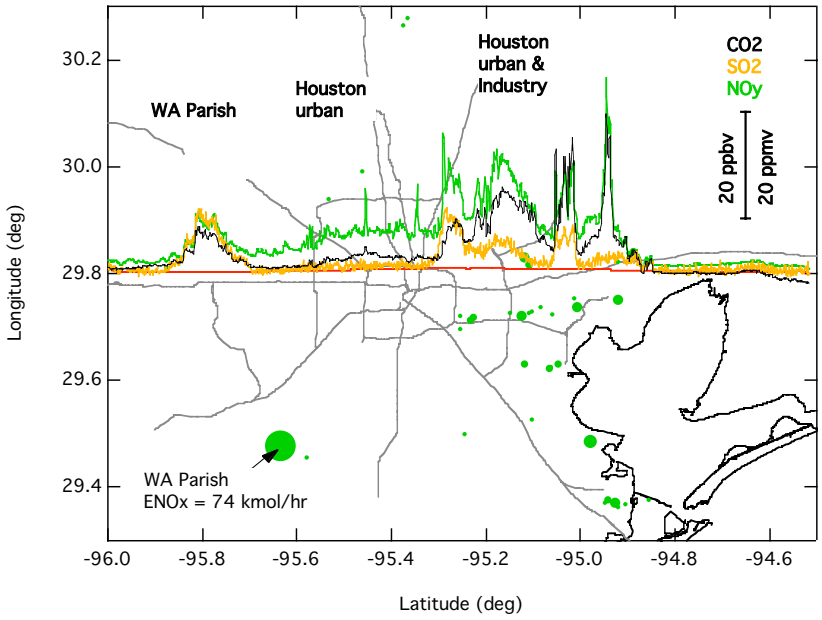

Figure 5. Shown is the flight track (red line) of the NCAR Electra across the Houston metropolitan area on 28 August 2000 when the winds were from the southeast. Superimposed on the flight track are the observed enhancements in the mixing ratios of CO_2 (black, 20 ppmv), SO_2 (yellow, 20 ppbv), and NOy (green, 20 ppbv).

Houston, the urban core contribution to the SO_2 and CO_2 emissions, in contrast, are small compared to the power plant and industrial emissions. This transect illustrates that most of the CO_2 in the Houston airshed is emitted by point sources – i.e., power plants and industrial sources.

In 2000, these point sources also accounted for a large fraction of the NO_x and most of the SO_2 emissions. However, since the 2000 study, the NOx emission scenario has changed very significantly. For example, the 2004 EPA point source emission inventory indicates that the NOx emissions from the WA Parish power plant have been reduced by a factor of 6-7 in comparison to 2000. Similarly, the NOx emissions of the major point sources along the industrial ship channel region have also been significantly reduced since 2000. These reductions need to be documented by the 2006 measurements and their implication for the formation of ozone and aerosols in the Houston area assessed.

3. What are the Volatile Organic Compound emission rates from petrochemical industries?

The TexAQS 2000 Study identified the important role that reactive light alkenes play in the rapid and efficient formation of ozone in the plumes emanating from petrochemical industries in Houston. The measurements showed that the light alkene emissions from some petrochemical facilities were severely underestimated. However, the nature and the relative importance of the sources – relatively continuous fugitive emissions or highly variable emissions accompanying flaring or accidental releases – still must be quantified.

The processes responsible for the emissions need to be understood before emission factors can be developed that can be included in regulatory and chemical forecast models.

In the TexAQS 2000 studies airborne measurements downwind of isolated petrochemical complexes were used to determine the absolute emissions rate of NOx and the emission ratios of hydrocarbons to NOx. While the emission rate of NOx inferred from these measurements compared well with those reported by the state of Texas, the ratios of ethylene and propylene to NOx were higher by more than an order of magnitude than those included in the State emission inventory. This was observed on all flights that measured in these emission plumes. While the hydrocarbon measurements were based on a few individual canister samples taken during plume intercepts, measurements of reaction products that were made at a higher temporal resolution confirmed that the high hydrocarbon to NOx emission ratios were representative. Measurements of the reactive hydrocarbons at a few seconds time resolution will be required during the TexAQS2006 study to improve the estimates of the hydrocarbon to NOx emission ratios. This will be of particular importance in the spatial allocation of the hydrocarbon emissions in the Houston Ship Channel region, with its complex agglomeration of a multitude of petrochemical facilities.

To address the important question of the temporal characteristics of the emissions from the petrochemical industries the use of ground based in-situ and/or remote sensing approaches should be investigated as a practical and cost effective alternative and supplement to airborne observations. Determination of the emission rates by several independent measurement approaches will build confidence and allow definition of the uncertainty in the current understanding of the emission rates of reactive hydrocarbons by petrochemical facilities.

4. Do ambient measurements of pollutant mixing ratios (e.g., CO/NOx, VOC/NOx, CO/CO_2, LAC/CO, NH_3/CO) reflect the temporal variation of urban emission ratios through diurnal cycles and weekday/weekend contrasts?

The weekly cycle of the human activity pattern in industrialized countries is reflected in lower NOx concentrations on weekends that have been seen at ground sites in urban areas (Marr and Harley, 2002, and references therein) as well by satellite measurements of the NO2 tropospheric column (Beirle et al., 2003). Urban emissions are a mixture from a variety of different emission sources that have specific diurnal and weekly activity patterns.

Traffic related sources have distinctive diurnal and weekly patterns. Gasoline powered vehicles emit CO, NOx, VOCs, CO_2 and aerosols. Diesel powered vehicles emit NOx, CO_2, aerosols, and in particular aerosols that contain light absorbing carbon (LAC). On weekdays, the emission from gasoline powered vehicles peak during the morning and evening rush hours, whereas truck traffic tends to peak during midday. On weekends the truck traffic strongly decreases, as does the morning rush hour peak of the gasoline-powered vehicles. Construction equipment that is mainly powered by Diesel engines is also mainly operated on weekdays. The stronger reduction of the Diesel engine emissions

on weekends compared to the gasoline engine emissions leads to a maximum in the VOC to NOx and CO to NOx ratios on weekends (Marr and Harley, 2002). As ozone formation is hydrocarbon limited in many urban areas, a reduction of the anthropogenic emissions that is accompanied by an increase in the VOC to NOx ratios can result in higher ozone concentrations in urban areas on weekends compared to weekdays (Marr and Harley, 2002, and references therein).

5. Can the direct emission of aerosols including black carbon be characterized by measurement of their ratio to major urban pollutants such as CO.

Baumgardner et al. (2002) have analyzed the relationship between light absorbing carbon (LAC) and carbon monoxide by comparing measurements from two sites in Mexico City and five urban areas in Germany. They found that the most important factors that affect the LAC to CO relationship appear to be the ratio of diesel to gasoline usage and the combustion efficiency of vehicles in a particular urban area. The LAC /CO ambient ratios can be compared to tunnel measurements of the emission ratios of the automotive source (Miguel et al., 1998). Furthermore, measurement of the LAC /CO ratio as well as the particulate to CO ratio at ground sites in the Dallas Ft. Worth and Houston airshed thus promises an important evaluation of the emission inventories.

Participants and Platforms:
NOAA and its extramural partners will instrument and deploy the NOAA WP-3D that will play an important role in emission verification. In addition, the NOAA Twin Otter lidar aircraft, the NOAA research vessel *Ronald H. Brown* and various instrumented ground sites and/or mobile sampling vans will be used to acquire data that can provide targeted emission verification for specific localized sources. The measured distribution of various trace gases will also be compared with satellite observations.

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 Twin Otter with the ETL lidar, and the R/V *Ronald H. Brown* along with several instrumented ground sites and satellites – 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 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. In addition to the use of the NOAA platforms, during the 2006 intensive we hope to make use of the composition of gas phase and aerosol distribution derived from retrievals based on satellite observations. Martin et al. (2004) have evaluated the GOME satellite measurements of tropospheric NO2 using the regional data from the TexAQS 2000 as well as the 1999 Southern Oxidants Study. A similar evaluation study will be of particular interest due the much improved spatial resolution of current satellite NO2 measurements as well as the significant reduction of the point

source NOx emissions that should be detectable by comparison of the 2006 and 2000 satellite measurements.

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 regressions 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 ±10%, providing an independent and accurate check on the ratio derived 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 variability, and to ensure that differential lofting of emissions (e.g., from multiple sources at different release heights) can be taken into consideration. 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. 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 compared.

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 flux 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 ± 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

Regional air quality and regional climate in Texas and the Gulf of Mexico are highly dependent on the transport and mixing of trace gases and aerosol particles. These processes occur on a variety of spatial scales, including atmospheric boundary layer (ABL), regional, intra-continental, and intercontinental. Boundary layer processes influence export to the free troposphere over the region. On the broader, continental 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 Africa is believed to have an impact on background particulate matter concentrations across the United States. The 2006 study is prepared to examine the full range of transport scales and their influence on regional air quality and climate in Texas. 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 impact Texas or are exported to downwind locations.

Two specific goals of the 2006 TexAQS /GoMACCS deployment are to further understand the meteorological and chemical processes leading to high-pollution events in the local Houston/Ship-Channel/Galveston-Bay area and to better understand controls on regional air quality, including factors affecting rural background levels of ozone, aerosols, and other pollutants. The local problem was already a major thrust of the 2000 project, and the regional focus is a new emphasis for 2006.

The sources of regional pollution include urban centers, industrial centers, automobile traffic, and power plants, which tend to be localized. Thus, the factors that control regional and background concentrations of pollutants are chiefly atmospheric transport processes acting on many length and time scales to carry pollutants and pollutant precursors away from the source areas. These length scales can range from local to intercontinental. For example, based on the case study day that produced the highest ozone concentrations during 2000, *Banta et al. [2005]* have shown that characteristics of the sea breeze at very local scales led to high ozone concentrations during the daytime, but nocturnal accelerations of the flow aloft could carry the pollution hundreds of kilometers from the sources overnight. The action of the nighttime transport thus transforms the localized daytime production of pollution into a regional transport problem, even during what might be considered "stagnation" or at least "light wind" conditions.

Science Questions

<u>Atmospheric-boundary-layer scale.</u>

Many aspects of the ABL behavior were found to be important for pollution accumulation and transport during 2000. Increased daytime mixed-layer depth over the Houston urban heat island and over the sea-breeze convergence zone carries pollutants to great heights during daytime (cf. Fig. 6); the suppression of vertical mixing over water (Galveston Bay, the Gulf of Mexico) keeps pollution concentrations high in the morning

offshore-flow layer; and in the evening surface cooling decouples the flow in the upper part of the previous-afternoon's mixed layer from surface friction. Such decoupling leads to nighttime acceleration of the winds aloft and thus becomes important for regional transport as noted. Contrasts in surface thermal properties, especially the land-sea contrasts, can generate significant horizontal variability in daytime mixing depths, including shallow mixing depths at the coastline that grow inland with distance from the shore during onshore flow conditions. These issues were identified during the drought conditions of 2000 using surface sensor arrays, ground-based remote sensing, and airborne measurements. The 2006 deployment should allow investigation of these processes under more normal (non-drought) conditions, and the addition of the *R/V Ronald H. Brown* and offshore profiler measurements will allow better specification of fluxes and ABL processes over water.

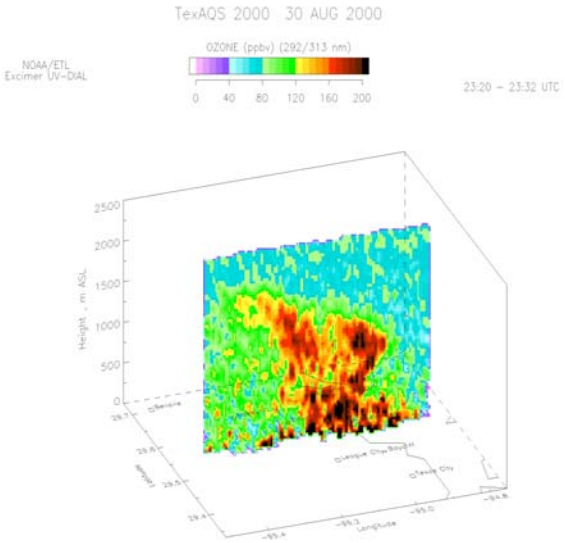

Figure 6: Time-height cross-section showing high ozone concentration air transported vertically within the sea breeze front at the western edge of Galveston Bay.

1. How do the heating and cooling cycle at the earth's surface and the resulting changes in ABL vertical structure affect horizontal transport from the major sources?

The daytime boundary layer over land is often well mixed through its depth, but if the air passes over a cooler water surface or when the land surface cools at night, the boundary layer develops layers of different properties, including speed and directional changes of the winds with height. Such shear can produce horizontal transport that varies strongly with height in the lowest 1-2 km above ground. When the underlying surface becomes warmer again, these layers generally mix out, which can result in fumigation of pollutant layers and increases in ground-level pollutant concentrations via downward mixing. Fully documented examples of such fumigation events are rare, and their overall impact on air quality needs to be evaluated.

2. What are the major processes that remove pollutants from the ABL?

Vertical ABL transport processes include exchange between the ABL and the surface via deposition and exchange between the ABL and the troposphere above the boundary layer.

The effectiveness of dry deposition processes at night is generally assumed to be small, but in fact is unknown. Examples of exchange between the ABL and free troposphere include cloud venting of boundary layer air, detrainment of boundary-layer air into the troposphere at discontinuities in the depth of the mixed layer, and entrainment of free tropospheric air that may contain pollution from distant sources or stratospheric O_3.

3. What is the horizontal variability of the boundary layer depth and what effects does this have on transport and mixing?

Differences in surface properties produce differences in mixed-layer depth *[Banta et al. 1998]*, such as the deeper boundary layer over the relatively warmer urban "heat island" *[Angevine et al. 2002]*, with the potential for pollutants to escape from the deeper mixed layer into the free troposphere above the boundary layer. This process has not been well documented in the atmosphere, nor has its potential for reducing pollutant concentrations in the mixed layer. When the daytime flow is onshore along the Gulf Coast, the mixed layer coming off the cooler water surface is shallow, but as it is transported inland, the mixed layer depth grows as an 'internal boundary layer.' Effects of this transition on vertical and horizontal transport are not well known.

Local scale.

High-pollution days occur preferentially during periods of light gradient, or large-scale, winds in southeast Texas. Such days are characterized by local sea-breeze development during daytime, followed by accelerated winds above the surface at night. Pollutants often accumulate in the vicinity of the sources during daytime and can be lofted deep into the PBL (c.f., Fig. 6). Studies have shown that high pollution concentrations are most often associated with a late-morning through mid-afternoon reversal of the near-surface winds at sites along the coast and just inland, as a result of the sea-breeze passage *[Darby 2005]*, with many days featuring pollutant buildup during morning and midday near the Houston urban-industrial sources, often followed by northward sea-breeze transport of the pollutants. Very high pollution events, however, occur when the sea breeze begins later in the afternoon and stalls, and a 1-3 hour stagnation period ensues just inland from the coast in mid to late afternoon *[cf. Banta et al 2005]*.

4. How do day-to-day variations in sea-breeze development (timing, intensity, etc.) affect peak local pollutant concentrations?

5. What is the extent of inland transport by the sea breeze, and what other factors control this transport?

A key factor controlling local pollution events in the Houston /Ship-Channel /Galveston-Bay area is the way in which the sea breeze develops on a given day. The strongest influence on the development of the sea breeze seems to be the large-scale, gradient wind vector, about which the diurnal sea-breeze cycle rotates *[Nielsen-Gammon 2002, Banta et al. 2005]*. The roles of other factors, which may include moisture and cloudiness, soil-moisture variation, lower-atmospheric stability, the depth of the sea-breeze flow layer,

etc., are not well known. The availability of offshore measurements by a wind profiler and the *R/V Ronald H. Brown* will be critical enhancements over 2000 for understanding the influence of the sea-breeze cycle on local-scale pollution.

The relationship between sea-breeze development and high pollution days also include a number of specific issues. Days with a reversal from morning offshore flow to late-morning or mid-afternoon sea-breeze flow are associated with high pollution, but do *all* days with a reversal have high pollution concentrations somewhere? As just described, the resultant sea breeze behavior as measured at a site near the coast is dependent on the "large-scale" wind, about which the diurnal sea breeze cycle rotates, but what is the appropriate measure of this large-scale wind, and can it be determined in advance for forecasting purposes and numerical model assessment? Additionally, what is the lingering effect of the previous day having been a high-pollution day, and how does this effect combine with the influence of the sea breeze?

Regional scales.

Regional processes affect both smaller regions, such as Eastern Texas, where rural pollutant levels are increased by direct transport and mixing from the major sources, to larger regions, such as the south-central United States (Texas, Louisiana and the northwest Gulf of Mexico), where intra-regional export, import, or recirculation of pollutants are important issues. Regional scale transport poses three important questions.

6. **Under what conditions does local urban pollution have a significant impact on other areas in the region?**
7. **How effective are nocturnal transport and subsequent daytime vertical mixing in dispersing pollution from the major sources into the surrounding region?**
8. **Are fugitive emissions from oil production important for regional pollution episodes?**

Under the daytime sea-breeze-reversal scenario just described, the lofting of pollution into the ABL results in deep columns of pollution by sunset, which are then distributed over the surrounding countryside by the accelerated nocturnal winds aloft (Fig. 7). Thus, as described previously, these conditions produce local pollution during daytime, but also contribute to regional pollution as a result of nocturnal transport. These types of day often cluster together into multi-day episodes, during which rural background concentrations of pollutants also increase, as a result of the nocturnal transport from the source regions and the overall relatively weak background winds. An issue for episodes is whether pollutants are carried out of the region or whether the sea breeze and other regional

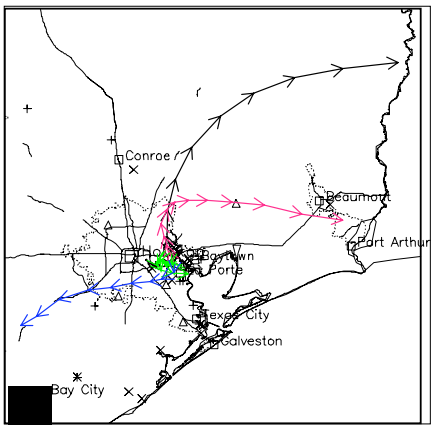

Fig. 7. Transport of local pollution during nighttime hours.

winds can re-circulate the pollutants and combine them with pollutants from other sources along the Gulf Coast.

Houston-area sources can also contribute to regional pollution when conditions are unfavorable for sea-breeze formation, as when boundary-layer winds are stronger (5-8 m s^{-1} or so). The wind speed and direction are more regional and persistent, and the diurnal sea-breeze cycle is at most a minor perturbation on the overall wind field. Winds of this velocity in the atmospheric boundary layer (ABL) produce a more plume-like behavior of the pollution, as shown in the airborne ozone lidar maps in Fig. 8. Despite the stronger winds, these conditions can still produce high hourly-averaged ozone concentrations, although overall concentrations are lower than in the previous scenario because of wind-speed dilution effects. Ozone and aerosols along with their precursors are transported over the countryside both day and night, and relevant issues are, whether the flow above the surface accelerates significantly at night, and whether the pollution continues to be carried out of the region.

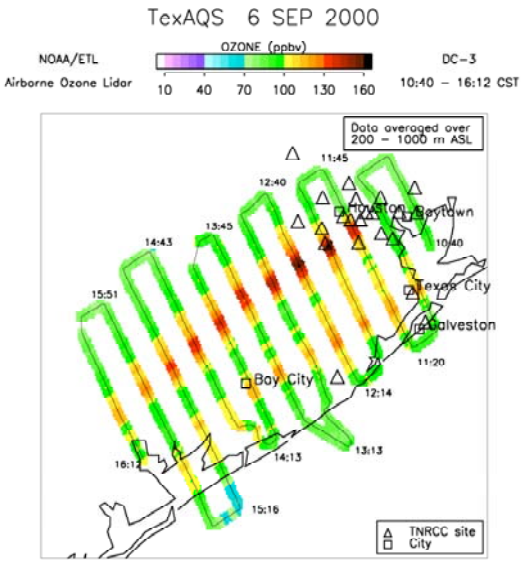

Figure 8: Flight track of ozone plume tracked downwind from Houston during the 2000 experiment. Ozone values are averaged over the layer *between 200 and 1000 m AGL*. Flow is from northeast to southwest. One site southwest of Houston registered hourly ozone concentrations of > 125 ppb.

Long-range, continental scales:

Long-range transport has implications for both air quality and climate. Several long-range transport issues can be identified that relate to the export of ozone and aerosols and their precursors from Texas to other regions of the United States or elsewhere, or to the import of the species into Texas from distant sources.

9. Do conditions exist when specific areas in Texas represent a source of pollution to other areas of the United States?

Can pollution originating in Texas remain at high enough concentrations to affect areas such as Atlanta, Chicago, the Ohio River Valley, or the East Coast of the United States? Can this pollution be carried northward to become entrained into the Great Plains nocturnal low-level jet and be incorporated into long-range transport (continental scales)?

10. What conditions exist when elevated background concentrations imported from distant source regions outside of Texas impact air quality or climate in the state?

Candidate distant source regions to affect Texas might include the Los Angeles area, Mexico, Central America, or recirculation from the southeast United States. Forest fires and other biomass burning were observed to affect Houston air quality during 2000, and smoke plumes from boreal fires in Canada and Alaska, tracked with satellite imagery, were observed above the northern Gulf Coast during July 2004. Saharan dust could add to the aerosol burden of the air over southeastern Texas. The impact of these distant sources on Texas air quality should be measured and the conditions attendant to them identified.

APPROACH

Participants and Platforms

NOAA researchers and extramural collaborators will address transport and mixing questions with arrays of instrumentation systems to measure the time-dependent 3-D distribution of meteorological and chemical quantities. A mesonet of surface stations will augment existing measurements sites to measure meteorological variables and chemistry at ground level, and some sites will be equipped to measure fluxes and surface energy budget components. Information on the vertical structure of these quantities will be documented by both ground-based and airborne instrumentation systems. Vertical profiles of wind and temperature, plus mixing height, will be provided by surface-based arrays of radar wind profilers with RASS, serial rawinsonde ascents, and a Doppler lidar, which will measure the fine-scale structure of the boundary layer either on the ship or at a land site. NOAA airborne platforms, including the WP-3D and the airborne ozone DIAL on a Twin Otter, will provide important information on the vertical and horizontal distribution of pollutants for forecast and model verification. Two other important capabilities are over-pressured "smart" balloons and the *R/V Ronald H. Brown*, which will carry surface-flux and energy-balance measurement capability, a Doppler wind profiler, an O_3 profiling lidar, a sophisticated complement of air chemistry sensors, and other instrumentation.

Numerical modeling. Numerical weather prediction (NWP) models are an important tool for diagnosing or predicting the *effects* of transport and mixing, and a number of modeling groups will participate in the execution and post analysis of the 2006 campaign. Important questions are, when can model results be used with confidence to predict transport of pollutants, what processes need to be better represented to improve these transport predictions, what is the nature of the limitations and uncertainties that exist in the model predictions, and what is their impact on predicted transport? Assessing model performance is a major thrust of TexAQS in 2006 and the subject of a later section of this document. These models also have the ability to be used as an effective tool to predict horizontal and vertical transport, improving the deployment and staging of mobile measurement platforms in 2006.

Deployment strategy

Measurement Strategy: The strategy for addressing the science questions is listed below as related to each scale of interest.

Atmospheric boundary layer scale: ABL-scale science questions will require knowledge of surface variables and fluxes and vertical profiles of many quantities. The vertical structure of the ABL and its relationship to surface heating and cooling will be addressed by the surface mesonet array, the profiler/RASS array, serial rawinsonde ascents, the Doppler lidar, and measurements on the *R/V Ronald H. Brown*. Airborne measurements will also provide vertical profile information, as well as other important spatial-distribution data to address transport effects and removal processes. Horizontal variability of the daytime mixing height and other quantities will be measured by airborne instrumentation and the profiler array.

Local scale. The same complement of instrumentation as used for the boundary-layer measurements will address the local-scale research questions. Especially important for defining the characteristics of the local sea breeze and its effect on pollutant distribution will be the airborne measurements, including the airborne DIAL and the WP-3D. The Doppler lidar provides important information on the fine-scale timing and vertical structure of the sea breeze flow for characterization of the sea-breeze behavior and for numerical model validation. The offshore measurements will be an important enhancement to the instrumentation deployed during 2000.

Regional scale. The focus on regional transport and regional air quality in 2006 will require a careful assessment of the effectiveness of various meteorological processes to transport pollutants from the major source areas to other areas of interest. One of the major questions is the role of nocturnal transport. *Banta et al. [1998]* report successfully using trajectories from hourly profiler-array data to find urban ozone blobs after overnight transport. For 2006, profiler data will be available in near real time, and a trajectory-calculating tool will be available for analysis, also in near real time. This resource should be used to guide sampling aircraft, including the airborne ozone DIAL and the WP-3D, during nighttime and early-morning flights to find the pollution layers and to characterize the regional transport and the fate of pollutants from the previous day. The airborne DIAL would probe the location, extent (horizontal and vertical), and properties of the O_3 distribution (diffuse or 'patchy'), and the WP-3D measurements would be used to confirm the origin and characterize the photochemical age of the air mass. On occasions when pollution is diagnosed as being transported to the north-northwest, the CMDL data from the tall tower near Waco, Texas, should be consulted to verify and characterize the transport. Analyses of these data sets will be used to assess the ability of NWP forecast models to properly and accurately represent regional transport processes.

Another tool that should be very useful in investigating regional transport is the over-pressured 'smart' balloon, which would be provided by extramural collaborators. These could be released close to the source regions at a representative altitude of interest to

track the overnight progress of the pollution, to confirm the profiler trajectories, and to aid in guiding the aircraft to the pollution. These platforms could be especially useful in multi-day episodes, to determine whether pollution originating in the Houston area can become recirculated in the Gulf Coast region or whether it is carried out of the region even during the episode.

Continental and larger scales: Regional- to hemispheric-scale transport models such as FLEXPART will be run in forecast mode during the experiment to identify pollution plumes transported within the south-central United States as well as polluted air masses being imported to the region. The NOAA aircraft and *Ronald H. Brown* can then be positioned to intercept these plumes. Furthermore forecast forward trajectories can be run from a flight track as soon as the flight is over to indicate the location of the sampled air masses over the next several days. The aircraft can then target these trajectory locations on subsequent days to create a regional scale Lagrangian study of the chemical and dispersive evolution of polluted air masses.

For long-range transport studies the NOAA aircraft will be able to wait for polluted air masses that are forecast to originate from other regions of North America or even Africa to be imported into the region. However, the NOAA aircraft will not have the range to effectively sample air masses far beyond the boundaries of the south central United States, unless suitcase flights are planned. The *in situ* measurement of polluted air masses that leave the borders of the south central United States will have to be made serendipitously by either stationary surface/tower instrumented sites, or other mobile airborne platforms. These additional mobile platforms include MOZAIC aircraft and NOAA CMDL aircraft. The European-funded Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program makes profiles of ozone and CO at airports in Houston, Dallas, Atlanta, Washington D.C., Chicago, Philadelphia, New York, Boston, Toronto, Montreal and Los Angeles. Although these profiles are not made on a systematic basis, 6-10 profiles are generally available somewhere over the United States for any given day, plus data at cruising altitude over North America and across the Atlantic to Europe. Also NOAA CMDL's new aircraft-based monitoring network will be partly available in 2006, providing additional trace-gas measurements up to 6 km at several locations across the United States on a weekly basis (the prospect of having these profiles made at a higher frequency during the study period needs to be pursued). Finally, remotely sensed trace gas measurements from polar-orbiting satellite instrumentation such as AIRS, MOPITT, SCHIAMACHY, or HRDLS (and other instruments on the new AURA satellite) can be used to track exported pollution plumes.

Chemical Transformation

Relevance

Chemical transformation is one of the principal processes controlling the abundance, distribution, and properties of ozone and aerosols in the atmosphere. As a consequence, chemical transformation affects their impact on climate, the environment, and 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 while the daytime chemistry that leads to ozone formation is qualitatively understood, nighttime chemical processes involving ozone and the importance of these processes is much less certain. On the other hand, the processes controlling aerosol formation and transformation during transport are even less well known.

Science Questions

The 2006 TexAQS/GoMACCS intensive will provide chemical transformation data that will help to address three overarching questions:

1. **What primary processes characterize the chemical evolution of ozone, aerosols and their precursors as they are transported from the source regions?**
2. **How do emissions from local and distant source regions interact to determine the air quality in Texas?**
3. **Do emissions in Texas influence the regional air quality and climate in distant regions?**

The challenge of the 2006 intensive will be to determine how gaseous and aerosol emissions in plumes from urban, power plant and petrochemical sources evolve chemically and physically as they are transported away from these source regions during the daytime and nighttime.

4. **What are the rate and efficiency of processes that control ozone and secondary aerosol formation during the daytime downwind of urban areas, power plants, and industrial sources?**
5. **How did the change in emissions from 2000 to 2006 affect the gas-phase and aerosol concentrations?**

Ozone is formed in the troposphere by photochemical reactions involving the oxides of nitrogen NO and NO_2 (summed as NO_x) and reactive volatile organic compounds (VOCs) [Haagen-Smit, 1952; Seinfeld and Pandis, 1998 and references therein]. Model studies have shown that O_3 formation rates and yields per NO_x molecule oxidized are dependent upon both the absolute concentrations of NO_x and VOCs and upon the ratios of these species [e.g., Liu et al., 1987, Derwent and Davies, 1994, Sillman, 2000]. Results from ambient measurements have confirmed that substantial differences in the rate and magnitude of O_3 production consistently occur in plumes downwind of different anthropogenic source types, characterized by different NO_x and VOC emission rates and the VOC/NO_x ratios that result [e.g., Ryerson et al., 1998, 2001, 2003; Daum et al., 2000; Nunnermacker et al., 2000; Kleinman et al., 2002]. In particular the measurements during the TexAQS 2000 study have shown that petrochemical industrial facilities can emit large amounts of highly reactive hydrocarbons and NO_x to the atmosphere; in the summertime, such collocated emissions are shown to consistently result in rapid and efficient ozone formation downwind [Kleinman et al., 2002; Ryerson et al., 2003; Neuman et al., 2002].

Along with the formation of ozone, particle growth in urban, power plant and industrial plumes has been systematically measured [Brock et al., 2002, 2003]. For example, during TexAQS 2000 plumes originating from the Parish gas-fired and coal-fired power plant, petrochemical industries along the Houston ship channel, the petrochemical facilities near the Gulf coast, and the urban center of Houston were studied (Figure 9). Most of the particle mass flux advected downwind of the Houston area came from the industries and electrical utilities at the periphery of the city rather than from sources in the urban core. In fact, clearly detectable gas-to-particle conversion was found only in plumes from large SO_2 sources. In SO_2-rich plumes that did not contain elevated concentrations of VOCs, the gas-to-particle conversion was consistent with the expected oxidation of SO_2. However, in plumes that were rich in *both* SO_2 and VOCs, the observed particle growth greatly exceeded that expected from SO_2 oxidation, indicating the formation of organic particulate mass (Brock et al., 2003). Finally, in plumes that were enhanced in VOCs but had little SO_2, and in the plume of the Houston urban center, no particle volume growth with increasing plume oxidation was detected. Since substantial particle volume growth was associated only with SO_2-rich plumes, these results suggest that photochemical oxidation of SO_2 is the key process regulating particle mass growth in all the studied plumes in the Houston-Galveston airshed. Clearly however, organic matter must contribute substantially to particle mass in petrochemical plumes rich in both SO_2 and VOCs. Quantitative studies of particle formation and growth in photochemical systems containing NO_x, VOCs, and SO_2, with improved real-time capability to measure particle composition, will be an important part of the 2006 study.

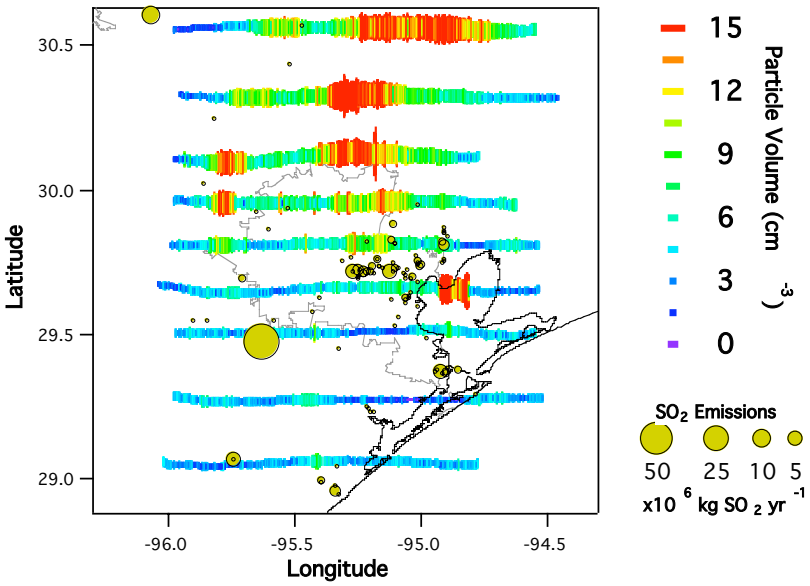

Figure 9. The figure illustrates the evolution of aerosols downwind of power plant, urban area and urban/industrial emissions that were observed during the flight of 28 August 2000. The locations of the SO_2 point sources are given by the yellow circles that are sized according to their emission strength as given in the legend.

5. Are the measurements of the products of VOC oxidation consistent with the current understanding of hydrocarbon chemistry?

The oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides leads to the formation of ozone in the troposphere. Besides ozone, other secondary species such as carbonyls and organic nitrates are formed that are characteristic of the parent VOC species. Ambient measurements of secondary species reflect the integrated effect of emissions, photochemical production and loss, as well as other removal processes. Analysis of ambient measurements of select secondary species can be used to evaluate the understanding of tropospheric photochemistry (Wert et al., 2003; Roberts et al., 2001, 2003).

In addition to the oxidation of gas phase VOCs, the investigation of the very sizable fraction of volatile organic compounds in aerosols is largely unstudied but of great importance in assessing the processes that determine air quality and climate. Measurements aimed at observing these chemical oxidation processes will be undertaken during the 2006 study.

6. What is the contribution of nighttime chemistry to the oxidation of NOx and what are the consequences for the formation and removal of ozone and aerosols?

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 section on "Transport and Mixing" emphasized the importance of transport during the night on regional air quality and climate. Horizontal transport can be greatly enhanced while vertical mixing is greatly reduced. This means that plumes can remain relatively concentrated while they are transported over greater distances.

The 2006 study will address the importance of the nighttime oxidation as a loss process for NOx and investigate its influence on ozone production. Measurements over different source regions will provide important new information concerning loss mechanisms for NOx at night. However, understanding the chemistry over the continent during the night will present a significant challenge due to the vertical layering of the atmosphere. This vertical layering that is encountered during the night makes finding and tracking plumes at the surface or from an aircraft extremely difficult.

Participants and Platforms:
NOAA and its extramural partners will instrument and deploy the NOAA WP-3D that will play an important role in understanding chemical transformation in the region. In addition, the NOAA Twin Otter lidar aircraft, the NOAA research vessel *Ronald H. Brown* and various instrumented ground sites and/or mobile sampling vans will be used to acquire data that can provide useful ancillary information concerning the chemical transformations that occur in plumes emitted by specific localized sources. The measured distribution of various trace gases will also be compared with satellite observations.

Deployment Strategy:
Daytime Chemistry: To study the emissions from power plants, industrial facilities, and urban centers during the daytime the WP-3D will fly transects within the planetary boundary layer (PBL) approximately perpendicular to the mean wind direction, upwind and at several distances downwind of the pollution sources of interest. Figure 9 shows an example of such a flight track that was flown during the TexAQS2000 study to examine the emissions and their chemical evolution from Houston and the surrounding power plants and the petrochemical facilities (Brock et al., 2003, Ryerson et al., 2003, Wert et al., 2003). Occasional profiles are included to probe the vertical extent of the mixed layer and the contrast in chemical composition of the PBL and the adjacent free troposphere. To determine if a particular plume is well mixed throughout the PBL, a transect or part of a transect can be repeated at multiple altitudes within and just above the PBL. To examine a vertical enhancement of the height of the PBL by, for example, the urban heat island effect, a spiral profile in the center and outside of the urban plume can be included. Since multiple transects and vertical profiling require additional flight time, a better alternative for the documentation of the vertical extent of the plume and the vertical

mixing within the PBL would be the co-deployment of the aircraft with extensive *in situ* chemical probes together with the ETL Lidar aircraft. While the Lidar aircraft maps the physical dimension of the plume by the height resolved measurement of ozone and aerosols along the flight track, the WP-3D documents the detailed chemical composition. This strategy proved highly successful during several research flights during the TexAQS 2000 study. During TexAQS 2000, this deployment strategy has been used to study the emissions near the Houston, Dallas - Ft. Worth, and the power plants in northeastern Texas as well as south of Dallas. In addition during these plume studies the aircraft maps out the distribution of the biogenic emissions over the different regions of eastern Texas and their interaction with the anthropogenic pollution. The profiles upwind of the emission sources of interest determine the importance of regional transport of pollution in east Texas.

Nighttime Chemistry: In situ measurements of NO_3 and N_2O_5 have been for the first time deployed on board the WP-3D during the NEAQS 2004 study. Building upon this experience systematic plume studies during the TexAQS/GoMACCS 2006 study 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.

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 and 2004 demonstrates that nighttime emissions into the shallow boundary layer can be followed by ship borne measurements. For example, during TexAQS/GoMACCS in 2006 the RV Ronald H, Brown will be able to sample the emissions that the land breeze carries from Houston to Galveston Bay or from Texas City to the Gulf of Mexico. This does not have to be done in a truly Lagrangian fashion, but intermittent plume interception at various distances from the sources will allow investigation of the chemical state of the plume at different ages since emission.

Power plant: Airborne studies of the evolution of power plant plumes will focus on the oxidation of NOx 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. The challenge will be for an aircraft, such as the WP-3D with *in situ* instrumentation, to locate these plumes in the horizontal as well as the vertical.

Daytime remnant: A second focus on the nighttime study will be to determine the fate of remnant NOx that was emitted by urban areas or point sources during the daytime. As turbulent mixing decreases in the late afternoon vertical wind shear can contribute to the horizontal dispersion of NOx in these plumes. The section on “Transport and Mixing” outlines a strategy that predicts nocturnal dispersion and transport using trajectory calculations that are based on wind profiler observations. That approach may allow air parcels to be followed and mapped during the night. If so, this approach can be used to guide aircraft to the plumes even after several hours of nighttime transport.

Aerosol Properties and Radiative Effects

Relevance:
Accumulation mode ($\approx$0.1 μm < D_p < $\approx$1.0 μm) and coarse mode (D_p > $\approx$1.0 μm) aerosol particles scatter and absorb solar radiation affecting visibility (Malm et al., 1994) and the Earth's radiative balance (e.g., Rasool and Schneider, 1971; Charlson et al., 1992; Ramanathan and Vogelmann, 1997). These particles can also act as cloud condensation nuclei (CCN), thereby influencing the albedo (first indirect effect, Twomey, 1974), lifetime (Albrecht, 1989), precipitation (Warner, 1968; Rosenfeld, 2000) and extent (Ramanathan et al., 2001) of clouds. Aerosol concentrations and their optical and radiative impacts are particularly high in regions downwind of sources, where diurnally averaged clear sky surface forcing range up to 30 Wm^{-2} (Russell et al., 1999; Ramanathan et al., 2001; Conant et al., 2003). International field campaigns during the past nine years have studied aerosol properties and their direct radiative effects downwind of Eastern North America (1996 – TARFOX; 2004 – ICARTT), Southwestern Europe (1997 – ACE-2), Southeast Asia (1999 – INDOEX), and Eastern Asia (2001 – ACE-Asia). During these nine years of major field programs, our scientific tools for measuring aerosols and their radiative properties and our understanding of aerosol chemistry and transport and transformation processes have evolved tremendously (IPCC, 2001). Nevertheless, the current understanding of aerosol effects on climate, both with respect to their direct radiative impact and the multiple (and mutual) effects of aerosol on clouds, leaves us with many unanswered questions. This situation is exacerbated by the complexity and regional/temporal variability of aerosol chemistry that is the source of large uncertainties in the optical, radiative, and cloud nucleating properties of the aerosol. At this time aerosols pose the largest uncertainties in calculations of radiative forcing of the climate system (IPCC, 2001, Figure 2).

As part of TexAQS/GoMACCS 2006, we propose to study the processes controlling the formation, transport and transformation of aerosol particles (see section on Chemical Transformation above) and the effect of chemical composition and mass size distribution on the optical, radiative, and cloud nucleating properties of the aerosol. In addition, we propose to directly measure the clear–sky radiative impact of aerosol over the Gulf of Mexico. Below we separate our science questions into aerosol "direct" radiative effects in cloud-free conditions and aerosol "indirect effects" where the focus is on aerosol-cloud interactions.

Aerosol Direct Effect Scientific Questions: The aerosol-radiation research plan is focused on several scientific questions described below.

1. What are the regional scale aerosol optical properties in Texas/Gulf of Mexico under different meteorological conditions and how do these properties change with altitude, location, distance from their source, time of day, and with changes in meteorological conditions?

Strategy: *Measure/calculate aerosol properties under a variety of conditions (e.g. downwind of different point and regional sources; at various altitudes and distances from shore; at different times of day; under different meteorological conditions). Compare directly measured aerosol scattering, backscattering, and absorption coefficients with those calculated from the measured size distributions and chemical composition (local closure).*

Since 1995, many field experiments have focused on the characterization of tropospheric aerosol properties to improve estimates of aerosol direct radiative forcing of climate. Several of these experiments have been located in marine environments immediately downwind of known continental aerosol source regions and have involved intensive field operations with measurements from ships, aircraft, and coastal land sites. Measurements over the ocean are prompted by satellite observations of backscattered radiation that reveal persistent seasonal aerosol plumes downwind of continents and the relative ease of accounting for the surface albedo over a uniform, dark ocean compared to inhomogeneous, bright land surfaces. *In situ* shipboard measurements have played an important role in these experiments as they provide information about aerosol chemical composition, size distributions, optical properties, and mass loadings in the boundary layer (Quinn and Bates, 2005). This information is required to fully understand the impact of regional aerosol plumes on climate and air quality.

2. How well can chemical transport models define the regional aerosol distribution?

Strategy: *Compare measured aerosol chemical and optical properties with those determined from chemical transport models.*

Radiative transfer models are used to calculate regional aerosol radiative forcing. These models require as inputs regional aerosol optical properties (single scattering albedo, backscatter fraction, mass scattering efficiencies, the functional dependence of scattering on RH) and aerosol mass distributions calculated from chemical transport models. Aerosol sampling during TexAQS/GoMACCS 2006 will be used to validate and refine the ability of chemical transport models to define the three dimensional aerosol distribution in this region (see following section on Validation of Forecasting Models).

3. How does the aerosol chemical composition affect the humidity dependence of aerosol light scattering?

Strategy: *Compare directly measured f(RH) (extinction and scattering) with values calculated from aerosol size distributions and chemical composition. Compare f(RH) values during periods of different organic mass fraction. Compare aerosol extinction at the surface measured by several techniques.*

A major finding during NEAQS 2004 was that *f(RH)* (extinction and scattering) was a function of the POM mass fraction. This needs to be further studied under different emission and meteorological conditions. It will also be critical to explore organic speciation to determine what compounds are controlling this RH dependency.

4. What are the dominant aerosol chemical components affecting aerosol light scattering (haze)?

Strategy: *Calculate aerosol mass fractions and mass scattering fractions using multiple linear regression and Mie modeling approaches.*

Organic aerosols dominated the total sub-micron aerosol mass and light scattering in the marine boundary layer off the New England coast during the summer of 2002 (Bates et al., 2005). This situation is not characteristic of aerosols downwind of Asia or Europe (Quinn and Bates, 2005). An understanding of the dominant aerosol mass and scattering fractions is critical for determining the health and radiative (e.g. water uptake, cloud nucleating properties) impacts of the aerosol and to develop mitigation strategies if needed.

5. What is the amount of light absorbing aerosol over Texas and the Gulf of Mexico?

Strategy: Measure the absorbing component of the aerosol at surface stations and with airborne in-situ and remote sensing instruments.

The deployment of numerous instruments that will measure both aerosol chemical composition and optical properties will enable assessment of the prevalence of light absorbing aerosol in the region and its spatial and temporal variability. This will provide information on an optical parameter crucial for assessment of aerosol radiative forcing, namely the single scatter albedo.

6. What is the direct (clear-sky) radiative impact of the aerosols in the study region over of Gulf of Mexico?

Strategy: *Measure/calculate clear sky radiative forcing under a variety of conditions (e.g. downwind of different point and regional sources; at various altitudes and distances from shore; at different times of day; under different meteorological conditions). Compare aerosol optical depth spectra measured by sunphotometers (surface and airborne) and retrieved from satellite radiances. Compare radiative fluxes determined from shipboard, aircraft, and satellite sensors (clear sky column closure). Compare clear sky forcing derived from models using measured aerosol properties and flux radiometers. Compare single scattering albedo measured at the surface and aloft with that derived from airborne sunphotometer and solar spectral flux measurements. Integrate the results of these measurements/comparisons to assess the regional clear-sky direct radiative forcing (c.f., Figure 10).*

The radiative impacts of aerosols are particularly high in regions downwind of sources, where diurnally averaged clear sky surface forcing range up to 30 Wm^{-2} (Russell et al., 1999; Ramanathan et al., 2001; Conant et al., 2003). To date direct measurements of

clear-sky radiative forcing have not been made over the Gulf of Mexico. TexAQS/GoMACCS 2006 will provide an opportunity to combine radiative flux and aerosol optical depth measurements at the surface, within the atmospheric column and from satellite. In 2006 a number of satellite sensors will be able to provide aerosol data from space. To reap the maximum benefit from these satellite measurements it is critical that we test and validate these measurements with in-situ and lower atmosphere/surface based column measurements (Diner et al., 2004). These measurements during TexAQS/GoMACCS 2006 can be made under a variety of aerosol/meteorological conditions over the Gulf of Mexico. This will likely be the first multi-platform integrated validation study for many of these new sensors.

Participants and Platforms

NOAA and their extramural partners will instrument and deploy the *RV Ronald H. Brown* and one or two small aircraft that are devoted to aerosol radiation studies. The NOAA WP-3 will be devoted primarily to process studies during TexAQS/GoMACCS but will participate in some multi-aircraft/ship experiments. The *in situ* and suborbital remote sensing measurements will be guided in the field with a hierarchy of model products and satellite observations.

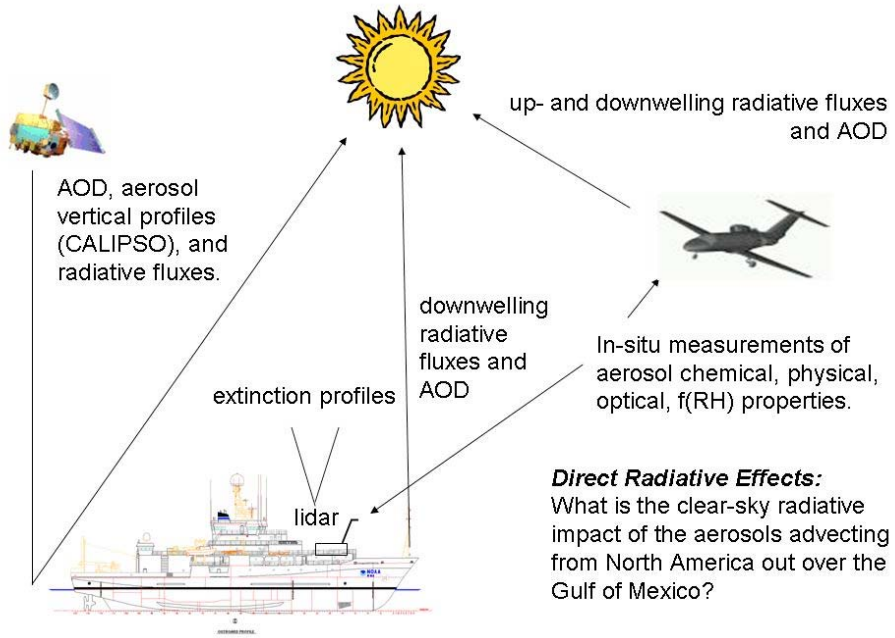

Figure 10. Schematic of platform deployment for measurements of the clear-sky radiative effect of aerosols.

Deployment Strategy (regional characterization and direct radiative forcing)

In situ and column measurements of aerosol properties will be made continuously aboard *Ronald H. Brown* in a variety of air masses to assess the effect of transport and transformation on these properties. Chemical forecast models and lidars (surface, aircraft and satellite (CALIPSO) will be used to determine the location and vertical distribution of aerosol plumes for targeted multiplatform column closure experiments. The ship and aircraft (c.f., Figure 10 and Figure 11) will be positioned to make *in situ* measurements (c.f., Table 1) of aerosol size distributions, chemical composition, *f(RH)*, and scattering and absorbing properties within the plumes at the surface (ship) and aloft (aircraft). Aerosol optical depth will be measured at the surface (ship), vertically resolved within the atmosphere (aircraft and ship lidars) and at the top of the atmosphere (satellite). In addition, downwelling radiative fluxes will be measured at the surface (ship), up- and downwelling radiative fluxes will be measured above and below aerosol layers aloft (aircraft) and upwelling radiative fluxes will be measured from space (satellite). Measurements will be coordinated with satellite overpasses (Terra, Aura, CALIPSO). 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 (Conant et al., 2003, Kahn et al., 2004; Schmid et al., 2003).

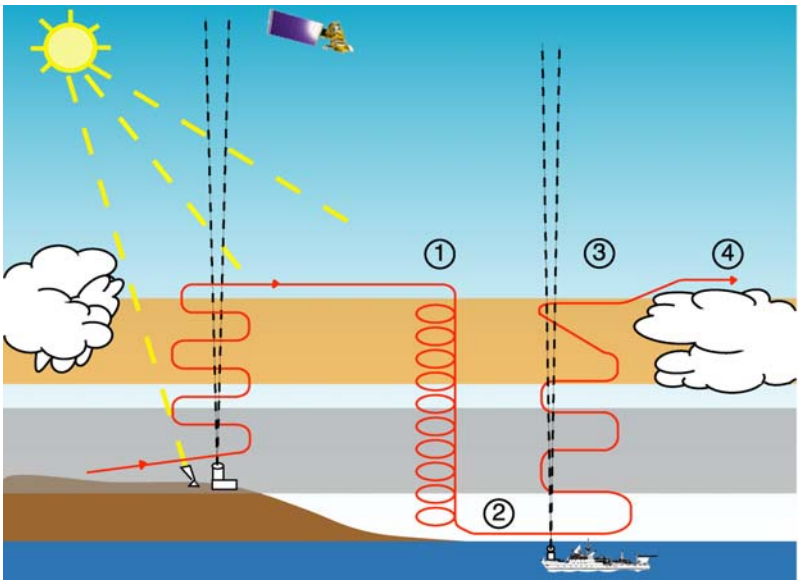

Figure 11. Illustration of the flight patterns used to reveal aerosol radiative effects and relate them to aerosol properties determined from space, air, land and sea. (1) Survey Vertical Profile. (2) Minimum-Altitude Transect. (3) Parking Garage. (4) Above-Cloud Transect.

The deployment strategy illustrated in Figure 11 can address the following experiments that will be required to (1) quantify aerosol properties and the direct aerosol radiative forcing and (2) intercompare and/or verify critical measurements:

- Measure/calculate aerosol properties and clear sky radiative forcing under a variety of conditions (e.g. downwind of different point and regional sources; at various altitudes

and distances from shore; at different times of day; under different meteorological conditions).

- Compare directly measured aerosol scattering, backscattering, and absorption coefficients with those calculated from the measured size distributions and chemical composition (local closure).
- Compare directly measured *f(RH)* (extinction and scattering) with values calculated from aerosol size distributions and chemical composition.
- Compare measured aerosol properties with those determined from chemical transport models.
- Compare aerosol extinction at the surface measured by several techniques
- Comparison of aerosol optical depth spectra measured by sunphotometers (surface and airborne) and retrieved from satellite radiances
- Compare radiative fluxes determined from shipboard, aircraft, and satellite sensors (clear sky column closure)
- Compare clear sky forcings derived from models using measured aerosol properties and flux radiometers.
- Compare single scattering albedo measured at the surface and aloft with that derived from airborne sunphotometer and solar spectral flux measurements.
- Integrate the results of these measurements/comparisons to assess the regional clear-sky direct radiative forcing.

Table 1. Proposed measurements and platforms for aerosol characterization and direct radiative forcing studies.

CATEGORY	**Parameters to Measure**	**Platform**		
		RHB	**Plane***	**Satellite**
Aerosol Chemistry vs Size	Anions and cations	X	O	
	Elemental carbon (EC)	X	O	
	Organic carbon (OC)	X	O	
	Organic carbon (speciated)	O		
	Organic function groups	O		
	Trace elements	X		
Aerosol Physical and Optical Measurements vs Size	Number vs size, accumulation & coarse modes	X	O	
	Absorption (spectral)	X	O	
	Scattering and backscattering (spectral)	X	O	
	Aerosol light scattering hygroscopic growth	X	O	
	Aerosol light extinction hygroscopic growth	X		
	Aerosol hygroscopic growth	X		
	Lidar backscatter profiles	X		
Radiation Measurements	Aerosol Optical Depth (spectral)	X	O	
	Radiative fluxes, solar and longwave, upwelling and downwelling (spectral)	O	O	
	Direct, diffuse, and total irradiances (spectral where possible)	O	O	
	UV flux	O	O	
Satellite-retrieved Fields of:	Aerosol Optical Depth (spectral)			Terra (MODIS, MISR)
	Radiative fluxes, solar and longwave			Aqua (MODIS)
	Surface albedo (spectral where possible)			
	Aerosol vertical profiles			CALIPSO
Meteorological Measurements	Wind speed, direction, RH, T, pressure, cloud type and amount, visibility	X X	O	

RHB – NOAA R/V *Ronald H. Brown*

PLANE – small plane devoted to aerosol radiation studies

X – indicates NOAA in-house capability

O – indicates needed measurement

* Note: the NOAA WP-3 will include complementary aerosol instrumentation (Table 4, page 49) that may be used in this study.

Aerosol –Cloud Interaction (Indirect Effect) Science Questions

The overarching science question to be addressed is:

How do the continental aerosols over the Houston metropolitan area and the Gulf of Mexico affect cloud microphysical and macrophysical properties and how do clouds affect aerosol size distribution and chemical properties?

The Houston metropolitan area is characterized by a range of aerosol types, as discussed in the previous section on "Chemical Transformations". The size distribution and composition of these particles are expected to vary greatly based on emissions, chemical processes (e.g., gas-to-particle conversion and heterogeneous processes), and transport (advection, venting, and fumigation). This range of aerosol conditions will provide a means of investigating the extent to which aerosol amount and composition effect cloud microphysics, and in particular cloud droplet number concentration. We will also explore the effect of aerosol on cloud lifetime. By addressing the following specific science questions our goal is to provide important information for evaluating the Twomey (1974) indirect effect, i.e., the effect of aerosol on cloud reflectance, and to reduce the uncertainty in cloud radiative forcing depicted in the IPCC chart (Figure 2).

Focused Science Questions:

1. How important is composition in determining the cloud condensation nucleating properties of an aerosol?

The literature contains a wealth of studies regarding the importance of aerosol composition, particularly with respect to inorganic, and more recently, water-soluble organic compounds. In addition, chemical effects on droplet activation such as nitric acid, surfactants, and organic films have been shown to be of potential importance (e.g., Nenes et al. 2002). The complexity of aerosol sources in the Houston area will afford an excellent opportunity to study the importance of composition vis-à-vis droplet formation.

- *Strategy: Compare measured CCN spectra with those calculated by thermodynamic models of aerosol activation, given measured aerosol size distribution and (size-resolved) chemical composition (CCN closure)*

The cloud condensation nucleus spectrum is an important property of the aerosol population and represents the number of particles that will grow to droplet sizes at a prescribed water vapor supersaturation. It is important to establish whether aerosol-CCN closure experiments can be achieved under a range of aerosol conditions, including those containing high organic aerosol fractions. Recent attempts at this closure (VanReken et al. 2003) during the CRYSTAL-FACE experiment were more successful than earlier attempts (e.g. Chuang et al. 2000) likely due to an aerosol composition dominated by ammonium sulfate, and improvements in CCN instrumentation. During the LBA-CLAIRE 2001 experiment, Rissler et al. (2004) predicted CCN concentrations typically to within 25% of the measured CCN, in a region where biomass-burning aerosols were

common. Organic species may represent a large source of uncertainty in closure studies (Charlson et al., 2001). Many aerosol-CCN comparisons implement Köhler theory and assume a mixture of a pure soluble salt, such as ammonium sulfate and insoluble material, neglecting detailed treatment of organic materials. Theoretical and laboratory studies indicate that organics may alter the activation characteristics of aerosol by reducing the mass accommodation coefficient of water (Bigg et al., 1986; Saxena et al., 1995; Feingold and Chuang, 2002) or by decreasing droplet surface tension (Facchini et al., 1999). Partially soluble aerosols (Shulman et al., 1996) and soluble gases (Laaksonen et al. 1998) may also contribute to uncertainties in the predictions of Köhler theory when the concentrations or properties of such species are unknown. The range of aerosol and gas-phase conditions in the Houston area will provide further challenges for CCN closure.

2. How well do our models represent aerosol activation in real clouds?

- *Strategy: Compare cloud drop number concentrations measured near cloud base with those predicted by models of cloud activation given observed updraft velocity and CCN spectrum (CCN-cloud droplet number closure). Assess the importance of chemical composition on CCN-cloud droplet number closure.*

Aerosol-drop concentration closure is similar in concept to aerosol-CCN closure except that model predictions of cloud drop concentration based on measured CCN (or aerosol size and composition) and cloud updraft velocity are compared to those measured directly by cloud probes. During the ACE-2 experiment, Snider and Brenguier (2000) predicted cloud droplet number concentrations from CCN measurements and the use of the Twomey (1959) parameterization and found them to be within a factor of two compared to measured concentrations. During the CRYSTAL-FACE experiment, Conant et al. (2004) found agreement between predicted (based on subcloud aerosol size and composition, and updraft) and measured drop number concentrations to ~ 15%. It is expected that closure is easier to achieve for particles of homogeneous and known composition, but is more difficult to achieve for heterogeneous composition, or in the presence of certain organic species. Closure is likely also easier to achieve in the unmixed cores of cumulus clouds such as those targeted by Conant et al. (2004). Closure may be more difficult in stratocumulus clouds, where the advection of drops may obscure the relationship between aerosol activation and drop number concentration.

An important part of this CCN-drop number closure effort is the measurement of the cloud updraft velocity that drives the production of supersaturation. For example, Leaitch et al. (1996) showed the importance of updraft in determining drop concentrations over the Gulf of Maine. The natural variability in updraft velocity and associated variability in supersaturation means that measurement of a supersaturation spectrum is of more importance in this closure effort than in the aerosol-CCN closure. The targeting of cumulus clouds with distinct updraft regions will facilitate this closure. Models will ingest aerosol size distribution/composition and vertical velocity measurements from aircraft and will compare measured and model-derived drop concentrations. Comparisons will be made both for individual clouds and in a statistical sense by using probability density functions (pdfs) of input parameters and comparing pdfs of model output with

pdfs of observed parameters.

3. How well do dynamical boundary layer models represent real clouds?

- *Strategy: Assess the ability of large eddy models to represent observed cloud microphysical processes, cloud dynamical and microphysical evolution, cloud lifetime and precipitation efficiency.*

The measurements to be acquired during TexAQS/GoMACCS 2006 represent an opportunity to test various models of aerosol-cloud interactions within a dynamical framework. Observations will provide constraints on boundary layer thermodynamic profiles, wind velocity components, microphysical properties, cloud depth, and precipitation formation. Visible satellite imagery will provide a geostationary view of cloud fields at 1-km resolution. Large eddy simulation models that integrate coupled dynamics, aerosol and cloud microphysics, and radiation (e.g., Feingold and Kreidenweis 2002), will be applied to a number of case studies. Model simulations will be used to test hypotheses pertaining to aerosol indirect effects, including aerosol effects on cloud fraction, cloud lifetime, and precipitation.

4. Can we detect evidence for cloud processing of aerosol and soluble gases?

- *Strategy: Look for evidence of cloud processing in observed soluble gas-phase species and aerosol size distributions in pollution plumes, up and downwind of clouds.*

Cloud processing of aerosol and gases can manifest itself in various ways, e.g., aqueous production of sulfate that creates a characteristic signature in the aerosol size distribution (Hoppel et al. 1991), perturbations in gas-phase chemistry as soluble gases are absorbed by clouds, removal of aerosol and soluble gases via precipitation (e.g., Respondek et al., 1995), and new particle formation in the vicinity of clouds (e.g., Hegg et al., 1990). Aircraft measurements upwind and downwind of cloud fields affected by pollution will be compared. When continental convective cloud formation is triggered by surface heating, pre-and post-cloud conditions will be compared. Convective redistribution of aerosol and gases by shallow cumulus clouds (a few km deep) will also be explored. Large eddy models will be brought to bear on these problems using trace gas and soluble-aerosol tracking tools.

5. Can we detect evidence for cloud suppression by absorbing aerosol?

- *Strategy: Identify the role of absorbing aerosols in reducing cloud development.*

Absorbing aerosol particles such as black carbon have been linked to reductions in cloud fraction and cloud liquid water path (Grassl 1975; Hansen et al. 1997; Ackerman et al. 2000; Koren et al. 2004) primarily through heating and stabilizing of the boundary layer. The importance of these absorbing aerosol particles is strongly linked to the vertical location of the particles (Johnson et al. 2004) and, over land, to the response of surface latent and sensible heat fluxes to the existence of the aerosol (Feingold et al. 2005).

During TexAQS/GoMACCS 2006 characterization of aerosol will include measurements of aerosol absorption profiles and calculation of associated heating rates. Assessments of cloud amount will be performed using remote sensing tools such as GOES 1-km resolution visible imagery. Other relevant measurements such as atmospheric temperature and humidity soundings will be used in the analysis. Large eddy models that represent the radiative effects of absorbing aerosol will be used to simulate case studies.

Deployment Strategy (indirect radiative forcing):

The primary focus for the TexAQS/GoMACCS 2006 aerosol indirect effect experiment will be on in-situ airborne measurements using a yet-to-be-identified small aircraft. The aircraft will be outfitted to provide a complete set of measurements pertaining to aerosol size, composition (inorganic and organic), hygroscopic growth and optical properties. Accurate characterization of cloud droplet size distributions will be a priority. Simultaneous measurements of updraft velocity and important meteorological parameters will be essential to this effort. Measurement requirements are listed in Table 2.

The small aircraft will perform independent missions associated with the science goals and will follow flight patterns that sequentially sample sub-cloud and in-cloud air. Specific flight plans will depend on the science questions being addressed. If possible, the small aircraft will coordinate with the NOAA WP-3D, which will have complementary aerosol and gas-phase measurement capabilities. This will enable stacked, column-type measurements, with the NOAA WP-3D focusing on characterization of sub-cloud aerosol while the small aircraft characterizes the drop size distributions. These stacked aircraft flights will simultaneously measure sub-cloud in-situ aerosol properties used to calculate CCN (physical size distribution, chemical size distribution, *f(RH)*), CCN, cloud droplet number concentration, drop size distributions, updraft velocities and liquid water content.

It is expected, based on prior climatological studies, that cumulus clouds will develop over the continent in association with daytime heating and moisture associated with the sea breeze. Flight planning will proceed accordingly and with consideration of the variability (in amount and composition) of local aerosol pollution sources. We will plan for overflights of the *R/V Ronald H. Brown* to assist with column closure experiments pertaining to aerosol direct radiative forcing and to target clouds over the Gulf of Mexico. Overflights of ground stations will be made as necessary. The preferred cloud-sampling mode will be a statistical one where multiple clouds at similar stages of development are targeted. Level legs below cloud and just above cloud base will be the preferred mode for the CCN and droplet number closure studies. Independent CCN closure studies will be made at surface sites such as the *R/V Ronald H. Brown,* since they do not require cloud sampling.

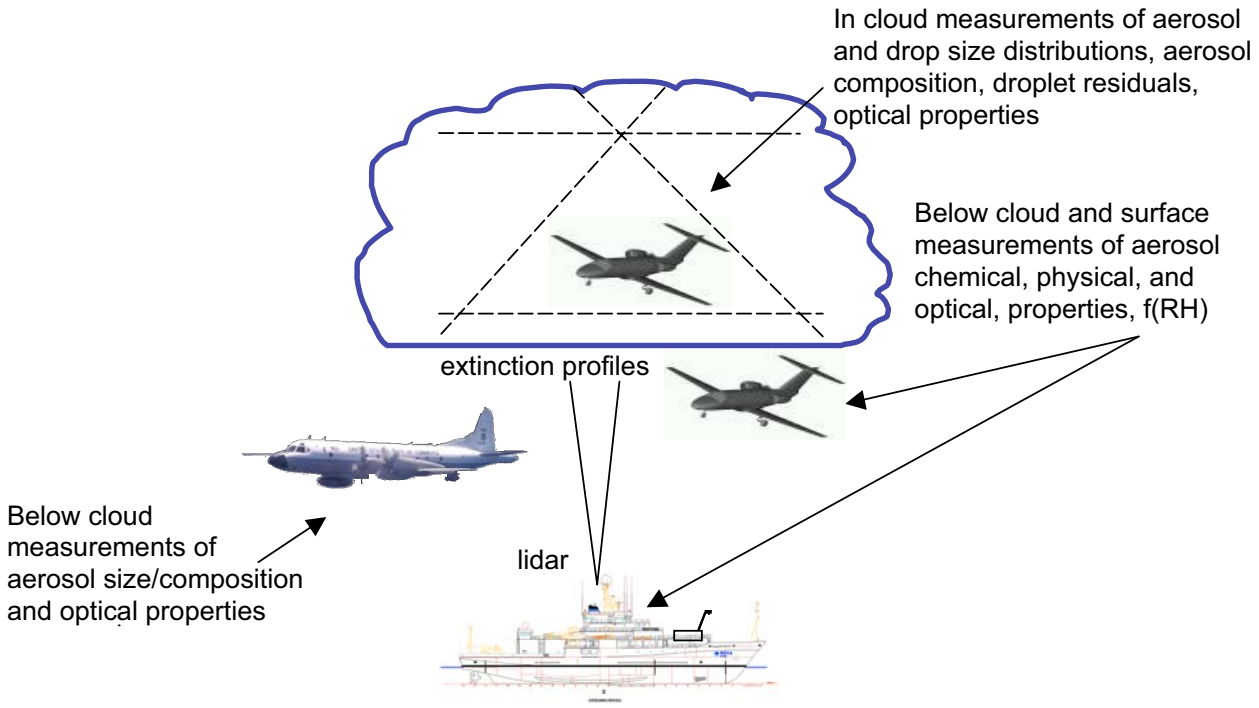

Figure 12: ***Schematic of deployment strategy for Indirect Radiative Effects.***

Table 2. Proposed measurements and platforms for aerosol indirect radiative forcing studies.

CATEGORY	**Parameters to Measure**	**Platform**		
		RHB	**PLANE***	**SATELLITE**
Aerosol Chemistry v Size	Anions and cations	X	O	
	Elemental carbon (EC)	X	O	
	Organic carbon (OC)	X	O	
	Organic carbon (speciated)	O		
	Trace elements	X		
	Organic functional groups	O		
Aerosol/Cloud Microphysical and Optical Measurements vs Size	Number vs size, nucleation, Aitken, accumulation &coarse modes	X	O	
	Absorption (spectral)	X	O	
	Scattering and backscattering (spectral)	X	O	
	Aerosol light scattering hygroscopic growth (f(RH))	X	O	
	CCN	X	O	
	Cloud drop size distribution		O	
	Cloud water content and surface area		O	
	Droplet residual properties (including chemistry)		O	
	Lidar backscatter profiles	X		
Radiation Measurements	Aerosol Optical Depth (spectral)	X	O	Terra (MODIS, MISR)
	Cloud optical depth		O	Terra (MODIS, MISR)
	Cloud reflectance		O	Terra (MODIS, MISR)
	Radiative fluxes, solar and longwave, upwelling and downwelling (spectral where possible)		O	Terra
	Direct, diffuse, and total irradiances (spectral where possible)		O	
	UV flux		O	
	Surface Albedo			Aqua (MODIS)
Meteorological Measurements	Wind speed, direction, RH, T, pressure	X	O	
	Cloud type and amount, visibility	X	O	
	Updraft velocity		O	

RHB – NOAA R/V *Ronald H. Brown*
PLANE – small plane devoted to aerosol-cloud-radiation studies;
* Note: the NOAA WP-3 will include complementary aerosol instrumentation (Table 4, page 49) that may be used in this study.
X – indicates NOAA in-house capability
O – indicates needed measurement

Evaluation of Forecast Models

Relevance

The effective management of the Nation's air quality requires a robust and reliable predictive capability. In addition, reliable air quality forecasts that provide sufficient warning of future adverse air quality provide a means for the public to minimize exposure. In 2004 NOAA initiated an air quality forecasting capability for the northeastern U.S. This system is being extended to the entire country. To be successful this system must 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 2006 field study, NOAA will deploy two prototype air quality forecast models that can provide operational air quality forecasts. The evaluation of these models with data collected in the 2006 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 the development of operational air-quality forecasting models, the study in 2006 will allow informal comparison of forecasts made by regional air-quality models with 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 2006 field study will provide diverse and rigorous tests of 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 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 2006 study.

1. How well can air quality models forecast air quality in Texas?

One of the important goals of the air quality component of the study will be to determine how well current state-of-the-art air quality models can forecast air quality in Texas. 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 2006 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 PM_{10} aerosol) over a large horizontal and vertical extent of Texas. This data set will therefore represent the centerpiece not only for model evaluation of ozone and its precursors, but for aerosols and visibility in Texas as well.

Model evaluations are 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 2006 evaluation study. There are fundamental differences in the basic formulation and meteorological foundations of the current operational and research air quality forecast models. Most models use off-line meteorology to drive pollutant transport, while other models such as the WRF-CHEM use online, or lock step calculation of meteorology and pollution transport. The effect these different model formulations have on Texas air quality predictions justifies a detailed statistical evaluation between the various models. Other important elements of 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 of the 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 put into quantifying emissions from thousands of sources, and eventually deriving an emissions inventory, are not sufficiently matched by resources or effort put 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 the study is the evaluation of emissions inventories on a relative as well as an absolute basis. Air quality forecast models comprise an important computational intermediates that relate the emissions inventories to atmospheric concentrations. This model evaluation study will provide the developers of the forecast models a clear picture of the ability of the models to capture both relative and absolute precursor abundances as they are reflected by and with in state uncertainties of the measurements. This allows 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 aircraft and ship-based studies of individual sources and urban regions planned for 2006 lend themselves directly to the evaluation of the photochemical and/or aerosol

mechanisms within each forecast model. The forecast models predict key oxidants (OH, O_3 in the daytime, NO_3 and N_2O_5 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 planned for the study (aircraft, wind profilers, ozone and aerosol lidars, and doppler lidar) 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 2004:*The air quality forecast model evaluation component of the NEAQS/ITCT 2004 study serves as a useful framework for model evaluation planning and deployment during 2006. Eight 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 O_3 measurements at several AIRNow sites, with O_3 and its precursors taken at the four University of New Hampshire AIRMAP ground sites and aboard the *R.V. Ronald H. Brown*, and with upper-air measurements from wind profilers and RASS temperature sounders at several locations. Time series of preliminary observations and model forecasts of O_3, CO, reactive nitrogen, and a number of surface and upper-air 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 as guidance for the deployment of aircraft and the *R.V. Ronald H. Brown*. This allowed the interception of urban plumes during the study period that proved useful for model evaluation.

In addition, the O_3 forecasts from the eight models available during the NEAQS/ITCT 2004 study were used to generate an ensemble O_3 forecast and a bias-corrected ensemble O_3 forecast that were also made available in near real time. Post-deployment analysis revealed that the ensemble forecasts of O_3, as well as the ensemble forecasts of $PM_{2.5}$, are statistically more accurate than for any individual forecast model. The TEXAQS/GoMACCS 2006 study would likewise realize this added benefit of having the best available air quality forecasts by centralizing the results of the forecast models available during the 2006 study period.

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/ITCT 2004 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 (NOx, CO, VOC, SO_2, PM_{10}, $PM_{2.5}$, and NH_3) be made available to those forecast and modeling groups involved in the study to insure compatibility between different models and different model resolutions. Close coordination with TCEQ and the University of Houston will be needed to adequately incorporate the best available emissions estimates for Texas into the various forecast models.

Regional Air Quality Forecast Models: The 2006 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 2006 to evaluate skill at predicting ozone photochemistry and PM_{10} and $PM_{2.5}$ aerosol concentration. Forecasts will be available at several model resolutions. Nested domains of 2 and 6 km centered over Texas within a 24 km grid covering the entire U.S. for the WRF-CHEM model are currently being considered. Forecasts from the NCEP/NWS model (either ETA/CMAQ or WRF-NMM/CMAQ) at 12 km resolution will also be available. The TAMU/UH (Texas A&M University/University of Houston) air quality forecast model would also be operational during the summers of 2005 and 2006. These three regional-scale forecast models are high priorities because of their spatial detail, the rigor of the basic physics and dynamics, and the importance they serve as operational or community based forecast models.

Additional Air Quality Models: At least two other regional scale air quality forecast models operational during ICARTT/NEAQS-2004 are also expected to be available during 2006. These include the BARON AMS, Inc. MAQSIP-RT model and the University of Iowa STEM regional model, which both operate multiple forecasts using different horizontal resolution. A number of hemispheric to global scale models that participated in ICARTT/NEAQS in 2004 are also expected to be operational during TEXAQS/GoMACCS in 2006. 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 Texas 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. These models also include Texas as a receptor region to various biomass-burning and anthropogenic sources occurring well outside the Texas study area. They therefore have the potential to serve as a valuable link in the comparison of regional versus long-range impacts to the visibility and air quality within Texas.

Model Evaluation: During ICARTT/NEAQS-2004, real-time comparisons of model forecasts were provided to ground-based and shipboard observers. Plans are being made to provide similar forecasts during 2006 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 Aeronomy Laboratory and the Environmental Technology Laboratory will provide the software, hardware, and personnel to assume these additional tasks.

The post-analysis of model forecasts is scientifically the most important aspect of the evaluation study. Model performance will be judged using a quantitative, statistical framework with

sampling throughout the entire ozone season . The air quality forecasts acquired during the study will be archived in a central location for convenient post-study analysis. NOAA will provide the software, hardware, and personnel to assume these additional tasks.

Mobile Platform Descriptions

The NOAA WP-3D Orion Aircraft

High pollution and haze events frequently affect Texas in the summer; however, the sources and the factors that shape the air quality are not well known. Both local and distant sources (transported pollution) are believed to play a role in these events. In turn, pollution from Texas can be transported out of state, with potential adverse impacts on regional air quality and climate.

An instrumented aircraft can uniquely address questions relevant to both climate and air quality components, e.g.:

- From the perspective of regional air-quality research, aircraft measurements can characterize multiple important features of the existing pollution issues in Texas. The 2006 study will address the processes of ozone and secondary aerosol formation, sampling both daytime and nighttime chemistry, and seek to improve the understanding of roles that emissions, chemistry and transport play in shaping Texas air quality.
- From the perspective of climate research, an aircraft can undertake a systematic study of the formation and evolution of the chemical and optical properties of aerosols from urban and industrial sources. The aim of such research is to address one of the more important open questions in climate research: how the various types of emissions and the subsequent atmospheric chemistry determine the optical properties of aerosols, and hence, the impact of these aerosols on radiative forcing in the atmosphere.

The NOAA Aircraft Operations Center (AOC) at MacDill AFB, FL maintains and operates NOAA's aircraft assets. Among these are the two four-engined turboprop aircraft Lockheed WP-3D Orions (see Figure 13). Since 1994, these aircraft have been

Figure 13: NOAA WP-3D Orion.

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 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 when fully instrumented	25,000 feet
Research speed	200 knots indicated air speed
Range	1600+ nautical miles
Fuel burn	4500 - 6000 pounds per hour
Fuel load	58,000 lbs, including reserves
Science payload	~5000 lbs inside fuselage; additional instruments in external wing pods

The operating range is ample to permit sampling of the primary pollution source regions, and to follow the transport and transformation of their emissions across Texas. Figure 14 shows the range of the WP-3D operating out of Ellington Field with a range of 700 nautical miles, assuming a return to Ellington. The above-cited operational range is an estimate and actual range is 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, i.e., with the fuselage loaded to capacity and additional instrumentation operated in external stores (pods) under the wings.

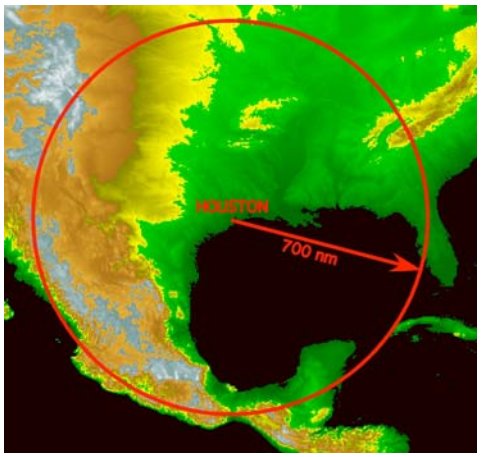

Figure 14. Operating range of the NOAA WP-3D Orion.

The WP-3D aircraft are operated by an AOC crew of seven (aircraft commander, pilot, flight engineer, navigator, flight director/ meteorologist, and two technicians) and can carry in addition several science personnel. The planned payload relies on having the full aircraft space and payload weight capacity available.

Instrumentation

Table 4 lists the proposed instrumentation package for the WP-3D during TEXAQS/GoMACCS 2006. Figure 15 illustrates the proposed payload.

Table 4: Proposed 2006 scientific payload for the NOAA WP-3D Orion aircraft.

Species	Instrument
Ozone (O_3)	NO/O_3 chemiluminescence
Nitric oxide (NO)	NO/O_3 chemiluminescence
Nitrogen dioxide (NO_2)	UV photolysis & NO/O_3 chemiluminescence
Total reactive nitrogen oxides (NO_y)	Au conversion & NO/O_3 chemiluminescence
Carbon dioxide (CO_2)	Non-dispersive infrared (NDIR) absorption
In-situ volatile organic compounds (VOCs)	Proton transfer reaction mass spectrometer (PTRMS)
Canister VOCs[1]	
Ethylene[1]	
Sulfur dioxide (SO_2)	UV pulsed fluorescence
Carbon monoxide (CO)	Vacuum UV resonance fluorescence
Formaldehyde (CH_2O)[1]	
PANs (PAN, PPN, etc.)	Chemical ionization mass spectrometer (CIMS)
HNO_3, NH_3	CIMS
Hydroxyl radical (OH), peroxy radicals (HO_2 + RO_2)[1]	
NO_3, N_2O_5	Cavity ring-down spectroscopy (CRDS)
Sulfuric acid (H_2SO_4)[1]	
Aerosol bulk ionic composition[1]	
Aerosol bulk composition	Aerosol mass spectrometer (AMS)
Aerosol number and size distribution (0.003 - 8 μm)	Nucleation mode aerosol size spectrometer (NMASS), laser optical particle counter (OPC), and white light OPC
Total (dry), sub-μm (as f(RH)) aerosol extinction	Cavity ringdown spectrometer
Dry sub-μm aerosol absorption (450, 550, 700 nm)	Particle soot absorption photometer (PSAP)
Fraction of absorbing aerosols	Soot incandescence
Cloud condensation nucleus (CCN) counter	CCN spectrometer
Actinic flux	Spectrally resolved radiometers
Broadband radiation	Pyrgeometers and pyranometers
Water vapor (H_2O)	TDL absorption
Air temperature	Platinum RTD
Dewpoint/frostpoint temperatures	Dewpoint/frostpoint hygrometer

[1] Desired but not yet specified.

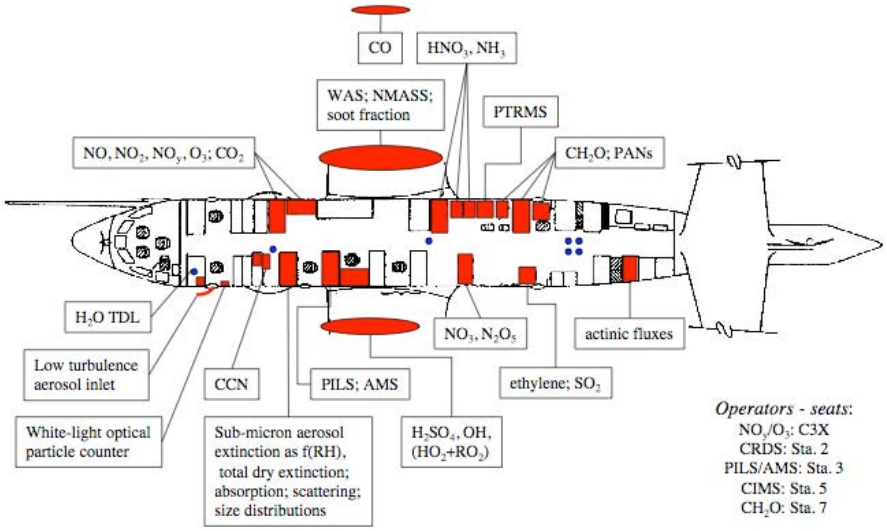

Figure 15. Proposed 2006 payload for the NOAA WP-3D Orion aircraft.

Operations

AOC operates the WP-3D aircraft under visual flight rules (VFR) and Instrument Flight Ranging (IFR) conditions, with some restrictions. As the external wing stores lack icing protection, flight into known icing conditions is not permitted. Minimum operating altitudes are determined by local flight conditions, but cannot override FAA regulations. Typical minimum daytime horizontal flight levels are 500 feet over land and 300 feet over water, depending on local air traffic and obstructions. We have in the past sampled over water for short periods at 150 feet. Over land, flying missed-approach patterns over local airfields permitted a temporary decrease in minimum operating height.

Typical Flight Planning Schedule

For the past eleven years NOAA has used a WP-3D aircraft extensively 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 PIs and the project science team.
- The flight planner reviews the 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 the 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 the final flight plan, and last-minute adjustments are discussed.
- After the 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 across the study region, to study dispersion and chemical and physical conversions within the plumes at 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 and patterns have been developed over the years and they will be used in the TEXAQS/GoMACCS 2006 study. Since several objectives and questions will be addressed in each flight, several strategies and patterns might be combined.

Coordination with Lidar aircraft

The capabilities on the *in-situ* (WP-3D) and the remote sensing NOAA Twin Otter (Lidar) aircraft strongly complement each other for many kinds of studies. 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 take advantage of having these two valuable resources available simultaneously.

NOAA Twin Otter

Overview

An important component of the 2006 experiment will be a differential absorption lidar (DIAL) deployed on a NOAA Twin Otter aircraft for remote sensing of local and regional ozone and aerosol distribution. Previous field studies have benefitted greatly from airborne measurements of ozone and aerosol profiles to characterize the three-dimensional structure of pollution plumes and measure variability in mixing layer height (Alvarez et al, 1998, Senff et al, 1998, Banta et al, 1998, Banta et al, 2005). 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 urban heat islands on pollution transport and mixing.

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

Aircraft Platform

The new ozone/aerosol instrumentation will be mounted on the NOAA DeHavilland Twin Otter. The NOAA remote sensing aircraft will tentatively be based at Ellington Air Force Base and will fly unpressurized at approximately 3-4 km above ground level for missions extending over approximately four to five hours. Relevant operating specifications for the aircraft during the experiment are expected to be as follows:

- Ground speed: 65 m s^{-1}
- Endurance: 4-5 hours
- Range: 550 nm
- Over-water capability
- Capability for multiple flights/day

On-board sensors

Flight plans for the Twin Otter will specifically address scientific objectives associated with the transport and evolution of pollution plumes, boundary layer structure, air quality forecasting, and intercomparison of observations. The primary instrument to be deployed on the aircraft will be a new down-looking ozone/aerosol DIAL system, which produces profiles of ozone and aerosol structure in the boundary layer and lower troposphere. The lidar system employs three tunable wavelengths in the ultraviolet spectral region between 280 and 300 nm. 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 the correction of potential errors in ozone calculations caused by aerosol backscatter gradients. Ozone measurements will be made at a horizontal resolution of approximately 600 m and vertical resolution of 90 m. Precision of the ozone measurement is expected to vary from

about 3 to 10 ppbV, depending on range and amount of intervening ozone. The longest of the three wavelengths at approximately 300 nm, which is least absorbed by ozone, is used to measure aerosol backscatter profiles, after correction for ozone extinction. Resolution for the aerosol measurements will be 600 m horizontally and 15 m vertically. Onboard the aircraft, a global positioning system provides a precise location for each lidar measurement, as for the plume tracking measurements in Figure 5 in the Transport and Mixing section. 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.

A key measurement objective for the Houston study will be the 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 16. 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 on the aircraft alongside the ozone lidar to measure surface skin temperature.

At this time, we are investigating the feasibility of incorporating a dropsonde capability on the NOAA Twin Otter. Dropsondes, particularly over the ocean, can provide information on wind structure associated with pollution layers, providing important information on the potential source of plumes observed distant from known sources. This decision will be based on the availability of funding and on assurances that reliable dropsondes can be procured.

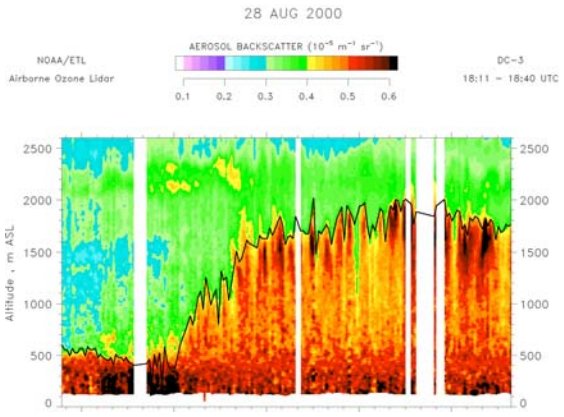

Figure 16: Vertical profile of lidar-observed aerosol backscatter showing a sharp change in mixing layer height at the Gulf of Mexico coast.

NOAA Research Vessel *Ronald H. Brown*

Transport of polluted air within the Gulf of Mexico plays an important role in shaping the air quality in coastal Texas. These same pollutants can also 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 Texas and long-range transport. Over the Gulf of Mexico 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 by the land-sea breeze circulation.

Figure 17. The NOAA Research Vessel *Ronald H. Brown*

An instrumented ship is an ideal platform to study the meteorological and chemical processes occurring off the coast of Texas and along the Houston and Galveston Ship channels. A ship can be used to sample polluted air masses as they move offshore or onshore and study the chemical transformations in the polluted marine boundary layer. Indeed, deployment of *R.V. Ronald H. Brown* (see Figure 17) during NEAQS 2002 and NEAQS/ITCT 2004 demonstrated unequivocally the value of this platform for providing unique sampling opportunities, unlike those from on-shore sites, which yield data that are frequently difficult to interpret due to contamination by local land-based sources, and unlike those from aircraft, which 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 5 (see also http://www.moc.noaa.gov/rb/index.html). The instrumentation (payload) capacity of *R.V. 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 17). Air samples are collected using towers or masts that extend 6-8 m above the deck (approximately 16 - 18 m 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.

Table 5. 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

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 Mexico. However, with an average cruising speed of 12 knots the ship is not a rapidly moving platform. 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.

The instrumentation proposed for deployment on RHB (see Table 6) 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 this field deployment 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 CO_2, and possibly O_3.

Table 6. Proposed instrumentation for deployment on *Ronald H. Brown*.

Parameter	Method
Photolysis rates (j-values)	Spectral radiometer
Ozone (O3)	UV absorbance
Ozone	NO chemiluminescence
Carbon monoxide (CO)	Nondispersive IR
Carbon dioxide (CO2)	Nondispersive IR
Sulfur dioxide (SO2)	Pulsed UV fluorescence
Nitric oxide (NO)	Chemiluminescence
Nitrogen dioxide (NO2)	Photolysis/chemiluminescence
Total reactive nitrogen oxides (NOy)	Au tube/chemiluminescence
Peroxyacyl nitric anhydrides (PANs)	GC/ECD
Alkyl nitrates (RONO2)	GC/MS
Nitrate radical (NO3); Dinitrogen pentoxide (N2O5)	Cavity ring-down spectrometry
Nitric acid (HNO3)	Mist chamber/IC
Water vapor (H2O)	Nondispersive IR
Continuous Speciation of VOCs	PTR-MS/CIMS
VOC Speciation	GC/MS
Formaldehyde (HCHO)	CHD fluorimetry
Radon (Rn)	Radon gas decay
Seawater/atmospheric CO2	Nondispersive IR
Enhanced measurement of radiative fluxes	Spectral radiometers
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)-IC
Aerosol Size and Composition	Aerosol Mass Spectrometer
Organic function groups	FTIR
Aerosol scattering (400, 550, 700 nm)	TSI Model 3563 Nephelometer
Aerosol absorption (400, 550, 700 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
Aerosol light extinction hygroscopic growth f(RH)	Cavity ring-down spectrometer
Total and sub-micron aerosol extinction	Cavity ring-down spectrometer
Ozone/aerosol vertical profiles	O3/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

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 2004 was very successful. This activity will be augmented in 2006 by more frequent communication between the ship and forecast personnel on shore. In addition extensive coordination between the ship and the various aircraft will be required. Efforts will be made to maximize opportunities for measurement 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 NEAQS/ITCT 2004, it will be expanded upon for 2006.

Figures 18 and 19 indicate the working area and intensive sampling area proposed for R.V. Ronald H. Brown during TEXAQS/GoMOACCS 2006.

Ship Sampling Objectives

There are four major scientific objectives, each associated with unique, but necessarily overlapping, sampling strategies.

Characterization of sources: Near-shore and ship canal 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). Also, special effort will be made to characterize marine vessel emissions (MVE).

Study of transport and transformation processes: A focus of the proposed research is the study of the chemical and physical evolution of polluted air masses in the Gulf of Mexico. When possible, plumes advected offshore will be sampled at successively longer distances downwind to examine chemical transformations related to plume aging in the MBL. Opportunities to sample well-aged plumes that have remained in the MBL for several days should be possible. Also, since significant chemical transformations occur at night, these studies of polluted air masses will occur during the entire diurnal cycle. Of particular interest is how the chemical and physical evolution of aerosols affects their optical properties.

Study of coastal impacts: Along-shore cruise tracks and transits up the Galveston ship channel are planned to characterize the effects of the recirculation of air masses by the sea-breeze/land-breeze circuit.

Study of radiative effects of aerosols: Cruise tracks are planned to examine aerosol properties in both polluted and clean conditions. In-situ MBL measurements will be

coordinated with aircraft measurements and satellite overpasses to assess the direct and indirect radiative effects of the aerosol.

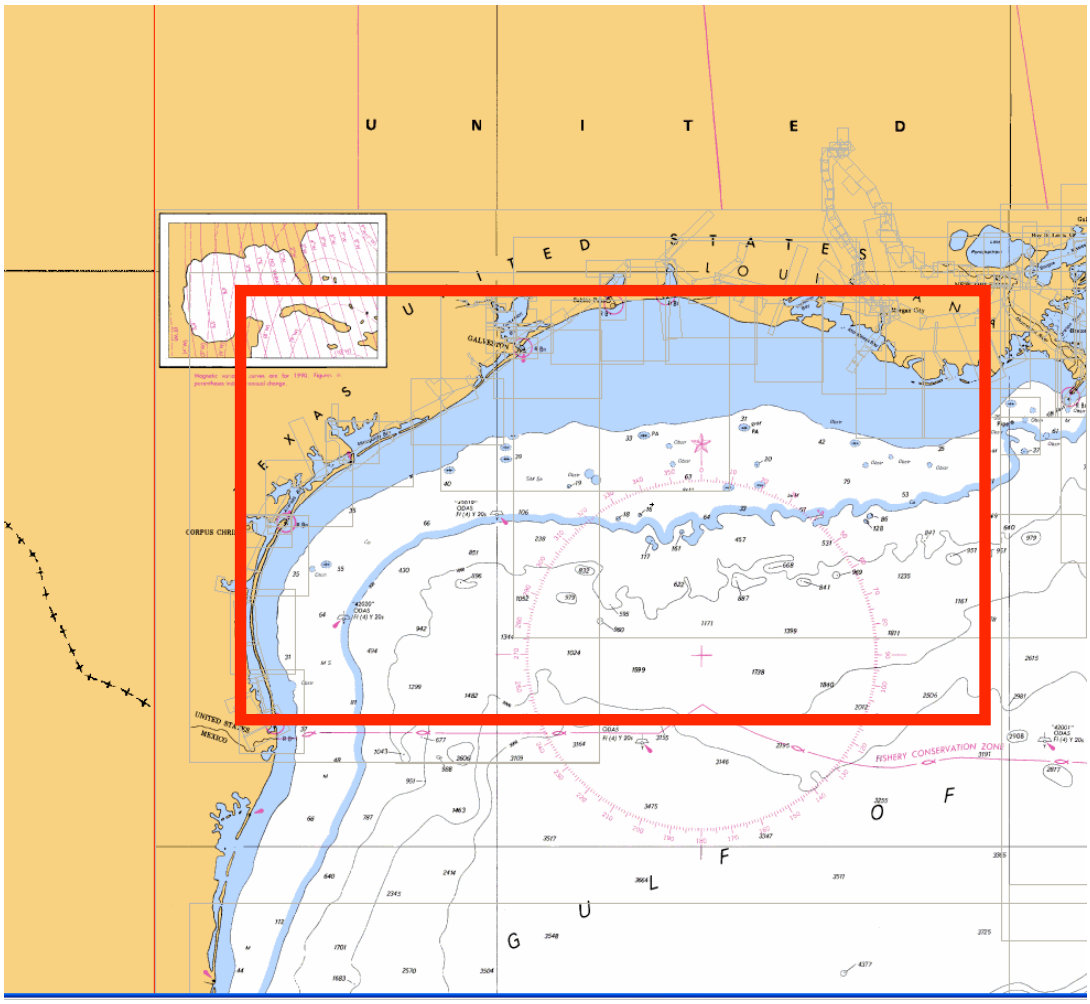

Figure 18. Ronald H. Brown working area in the Gulf of Mexico (within red square).

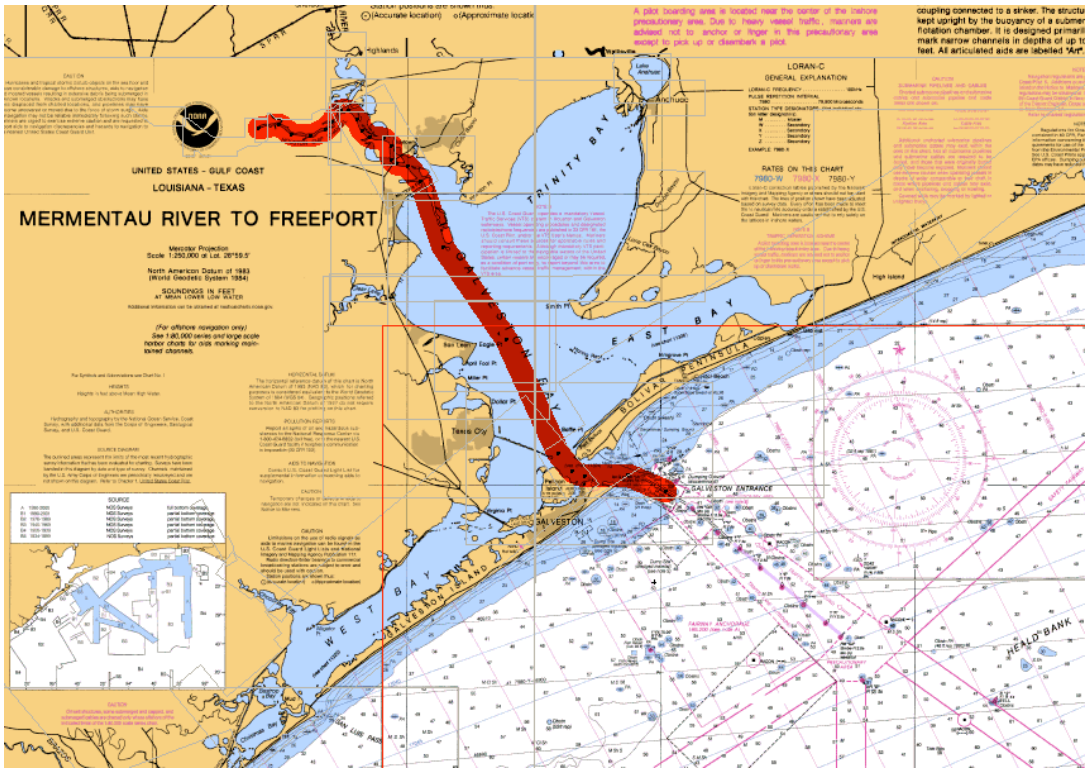

Figure 19. Intensive operations area showing ship track through Galveston Bay and up the Houston ship channel (heavy red line).

Other Measurements

Ground-based Measurements

Upper-air observations

East Texas experiences a complex diurnal cycle of meteorology caused by its proximity to the Gulf Coast, large urban areas and other heterogeneous land-surface types that produce local circulations, and varying synoptic regimes experienced during different seasons. A multi-season characterization of the meteorological processes controlling the stagnation and transport of atmospheric pollutants in and out of East Texas is, therefore, required in order to gain a better understanding of the region's air quality. The long-term meteorological measurements collected during TEXAQS/GoMACCS will also allow scientists to study how the mixing and transport mechanisms outlined in Section 2b respond to diurnal, seasonal, and annual cycles.

NOAA and partners in TEXAQS/GoMACCS will enhance the upper-air observing system in central and southeastern Texas by deploying seven integrated boundary layer observing systems for a ~1.5 year extended observing period starting in April 2005 (c.f., Fig. 20). Included in the plans for this network is a wind profiler deployed on an oil platform in the Gulf of Mexico. The enhanced profiler network was designed to capture important transport corridors within East Texas. The 915-MHz Doppler wind profilers in the enhanced network are of the type described by Carter et al. (1995). These instruments provide continuous profiles of wind speed and wind direction in the boundary layer and lower free troposphere and derived mixing heights. Each land-based profiler will also include a radio acoustic sounding system (RASS) for temperature profiling. The vertical coverage of the wind profilers is typically 120 m to 4000 m, depending on atmospheric conditions, and the profiles are sampled with either 60-m or 100-m vertical resolution. The vertical coverage of the temperature profilers is typically 120 m to 1500 m, but degraded performance can be expected in high wind conditions. In addition, four of the profiler sites deployed by the NOAA Environmental Technology Laboratory will include a global positioning system (GPS) receiver for integrated water vapor measurements and a 10-m tower for characterizing surface meteorology (pressure, temperature, relative humidity, wind, precipitation, solar and net radiation). Additional profiler deployments may be added for the intensive observing period in 2006. During this period, serial rawinsondes will be launched (by non-NOAA participants) in the Houston area at a site to be determined.

Hourly data from the existing and enhanced wind profiler networks will be available in real-time via web sites hosted by NOAA. An interactive web-based profiler trajectory tool developed by the NOAA Environmental Technology Laboratory will also be available to help scientists document transport in the region and to help in the planning and execution of missions for the mobile platforms during the 2006 intensive.

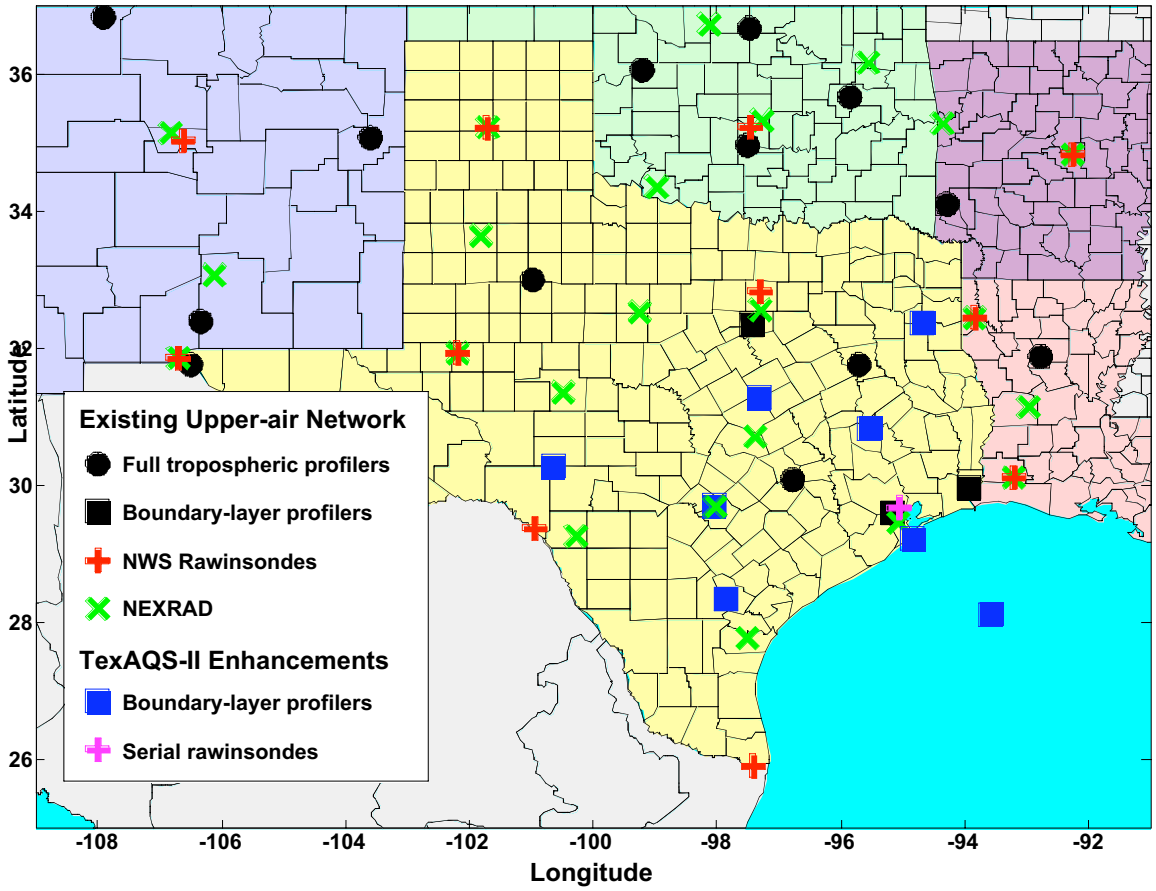

Fig. 20. Regional map showing the locations of upper-air observing sites in the existing operational network and enhancements to the network in central and southeast Texas that will be in place for TEXAQS/GoMACCS.

Satellite Observations

Background

Retrievals of aerosol and trace gas information from current research and operational satellites have great potential to assist in several of the TEXAQS/GoMACCS science objectives. Instruments on NASA and NOAA satellites are currently able to observe several of EPA's criteria pollutants (Table 7). While polar-orbiting satellites (e.g., MODIS) provide coverage once a day globally, geostationary satellites (e.g., GOES) provide coverage over the continental United States once every fifteen minutes. A multiple platform and sensor approach, integrating *in situ* and satellite data with modeling, might be essential to address TEXAQS/GoMACCS science objectives.

Table 7. A list of NOAA and NASA satellites and their measurement capabilities.

Satellite Platform: Web site	**Instruments**	**Some key data products**	**Vertical Resolution**
NASA Aura: http://eos-aura.gsfc.nasa.gov	TES OMI	CO, CH_4, O_3, HNO_3, NO_2 O_3, NO_2, SO_2, H_2CO, aerosol optical depth, aerosol type	Trop. column/4 km Trop. column
NASA Aqua: http://eos-pm.gsfc.nasa.gov	MODIS* AIRS* AIRS* AIRS*	Aerosol optical depth O_3 CO Aerosol optical depth	Trop. column UTLS Trop. column Trop. column
NASA Terra: http://eos-am.gsfc.nasa.gov	MOPITT MISR MODIS*	CO Aerosol optical depth Aerosol optical depth	Trop. column Trop. column Trop. column
NASA CALIPSO http://www.calipso.larc.nasa.gov	CALIOP	Aerosol backscatter ratio	Trop. vertical profile
NOAA GOES http://www.ssd.noaa.gov/PS/FIRE/GASP/gasp.html	Imager	Aerosol optical depth (30 minute interval)	Trop. column (land and water)
NOAA N16, N17, N18 http://www.osdpd.noaa.gov/PSB/EPS/Aerosol/Aerosol.html	AVHRR	Aerosol optical depth	Trop. column, (water only)
NOAA GOES http://www.orbit.nesdis.noaa.gov/smcd/emb/gsip/index.html	Imager	Shortwave flux (hourly)	Surface
NOAA GOES	Imager	UV (erythemal) flux (hourly)	Surface
NASA http://asd-www.larc.nasa.gov/ceres/ASDeceres.html	CERES	Shortwave and longwave flux	Top of the atmosphere; surface

*Available through NOAA in near real time

Although satellite data has some disadvantages compared with other means of observing ozone and aerosols, the advantages of including satellite information currently outweigh the disadvantages. Accuracies of satellite retrieved aerosol optical depths and trace gases are not as good as measurements made from ground because satellite retrievals tend to have higher uncertainties. These uncertainties are associated with converting slant column retrievals to column amounts and isolating the tropospheric column from the total column in the case of trace gases. For aerosol retrievals, difficulties in modeling aerosol type and variability in surface reflectance lead to large uncertainties. Nevertheless, while the ability to measure trace gases and aerosols at the desired spatial resolution, temporal resolution, and accuracy might not be realized for several years, the benefits of exploiting these measurements for air quality studies are so substantial that the validation required to exploit them should be pursued immediately. .

Satellite data of aerosols and trace gases have three potential applications for the TEXAQS/GoMACCS field campaign:

Using the satellite data in near real time:

- Image loops (especially from GOES) for aircraft/ship flight deployment
- Comparisons with *in situ* measurements
- Assimilation into forward trajectory models to forecast plume location

Retrospective looks at the data collected during TEXAQS/GoMACCS:

- Comparisons of satellite and ground/aircraft/ship based measurements of various parameters
- Validation of satellite retrievals using *in situ* measurements. Assessment of uncertainties in the retrieval algorithms due to various assumptions. Reprocessing of satellite data with assimilated field measurements
- Extending the spatial (horizontal) dimension for studying problems such as contributions of long range transport to local air quality

Using the satellite data in modeling studies:

- Verifying chemistry and transport model forecasts with satellite data
- Diagnosing sources of uncertainties in chemistry and transport models
- Assimilating satellite data to improve initial and boundary conditions in the models
- Radiative effects of aerosols

These applications will cover various TEXAQS/GoMACCS research topics as described below.

<u>Emissions Verification</u>

Satellite retrievals of CO, NO_2, SO_2, and H_2CO will be a good data resource for quantifying emissions from various sources in Texas. Studies are already underway to

determine isoprene emissions from OMI H_2CO data (Jacob et al., 2005). AIRS and MOPITT CO retrievals have thus far been primarily used to track plumes from biomass burning; plumes from biomass burning stay aloft and are easy to detect from satellites. It is unclear how useful satellite measurements of CO can be to study urban/industrial emissions if CO remains close to the ground. However, studies can be carried out to determine their usefulness, especially in combination of OMI's light absorbing aerosol index to characterize the source as diesel or gasoline. Satellite data have never been used to study processes at such small spatial scales, and TEXAQS/GoMACCS will provide that opportunity. For the satellite community, these studies can be very beneficial, as they will expose limitations in satellite retrievals and offer insights into needed improvements in sensor technologies and algorithms, so that these kinds of routine applications can be realized.

Long Range Transport

Satellite data have been very useful in diagnosing long-range transport, primarily because it occurs in free troposphere and is easily detectable by satellites. We would like to address the issue of entrainment of pollutants transported from remote sources into the boundary layer, and the impact on local air quality. NASA will be launching CALIPSO in June 2005, which has a lidar that can detect tropospheric vertical profiles of aerosol backscatter ratio. These measurements, combined with GOES, MODIS, OMI, and MISR aerosol optical depth retrievals, will be valuable in providing a three-dimensional look at pollution plumes. Additionally, integrating data from multiple sensors will optimize the information on aerosol type, and location in space (horizontal and vertical scale) and time (GOES aerosol observations have a refresh rate of 30 minutes).

Numerical Modeling

NOAA's National Weather Service has a mandate to issue nationwide hourly ozone (by 2009) and $PM_{2.5}$ forecasts (by 2014). The NWS has already begun issuing ozone forecasts for the northeast and conducting experimental $PM_{2.5}$ forecasts. It is currently using the Eta-CMAQ modeling system and will soon migrate to the WRF model with integrated meteorology and chemistry. Primary sources of uncertainty in model forecasts are uncertainties in intial/boundary conditions and emissions. Satellite data have the potential to improve forecasts by providing more precise initial/boundary conditions. *In situ* data collected during the TEXAQS/GoMACCS will also be useful in verifying forecasts and diagnosing various sources of uncertainties. In addition, a retrospective analysis of the TEXAQS/GoMACCS data from an integrated satellite-*in situ*-model approach will help to determine biases and errors in the air quality modeling system.

Aerosols and Radiative Forcing

Estimating the radiative impact of aerosols requires concurrent aerosol and radiation measurements. The spatial (and temporal) variability of both quantities is readily observed by satellites. Aerosol optical depth (the primary factor affecting radiative forcing) over the ocean has been retrieved operationally at NOAA/NESDIS for over two decades. For example, AVHRR observations have been used to estimate the aerosol indirect effect for summertime stratiform clouds in the Northeastern Atlantic (Matheson, Coakley and Tahnk, 2004). Instruments flown on NASA satellites have also been

providing aerosol data: TOMS has the ability to estimate absorbing aerosols, and the MODIS instrument is capable of estimating aerosol optical depth both over land and ocean in two different particle size regimes (fine and coarse modes). These measurements, coupled with top of atmosphere (TOA) and surface fluxes derived from the Clouds and the Earth's Radiant Energy System (CERES) instrument on NASA's EOS satellites, have been used to estimate the direct radiative forcing of aerosols in the shortwave and longwave part of the electromagnetic spectrum. In addition, satellite derived radiative fluxes can be used to constrain models at the upper and lower boundaries of the atmosphere for estimating the radiative impact of aerosols. However, before satellite measured fluxes can be used in assessing the radiative effects of aerosols they need to be thoroughly tested and evaluated with surface measurements; their consistency must also be characterized.

Validation of Satellite Retrievals using TEXAQS/GoMACCS Data

NOAA/NESDIS will use data collected during TEXAQS/GoMACCS in evaluating various assumptions made in our GOES and AIRS aerosol optical depth retrieval algorithms. The aerosol algorithms use look-up tables created using a continental aerosol model, for which the single scattering albedo is ~0.9. This includes assumptions about aerosol type, size distribution, and refractive index. However, studies have shown that there are significant variations in aerosol type and size distributions over the CONUS. These variations are largely dictated by sources of pollution (e.g., forest fires, urban/industrial, or dust). Ground-based and aircraft-based observations of aerosol parameters (size distribution, vertical profiles, etc.) will be used to build new aerosol models and create new look-up tables. Sensitivity studies will be performed to test the impact of measured aerosol information on GOES aerosol optical depth retrievals.

Infusing satellite Data into Environmental Applications (IDEA)

IDEA is a two-dimensional, near real-time system that integrates MODIS aerosol optical depth, $PM_{2.5}$ measurements, meteorological data, and models for use by EPA and state and local forecasters in monitoring and predicting $PM_{2.5}$ concentrations for public notification (Al Saadi *et al.*, 2005). IDEA was developed in a cooperative project between NOAA, EPA and NASA to provide real-time views of AOD from MODIS, compare these with the EPA AIRNow ground monitors, include trajectory information for forecast guidance, and provide a brief analysis for the public. Planning is underway at NESDIS to transition IDEA to NOAA to run in an operational (24-hour, seven-day) environment. This product, if up and running at NOAA by summer of 2006, will be a very useful forecast tool to coordinate ship/aircraft deployment. If it is not ready to become operational at NOAA by 2006, arrangements can be made with University of Wisconsin (U. Wisc) to provide IDEA forecast guidance. It is currently running at Wisconsin in a pre-operational mode.

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