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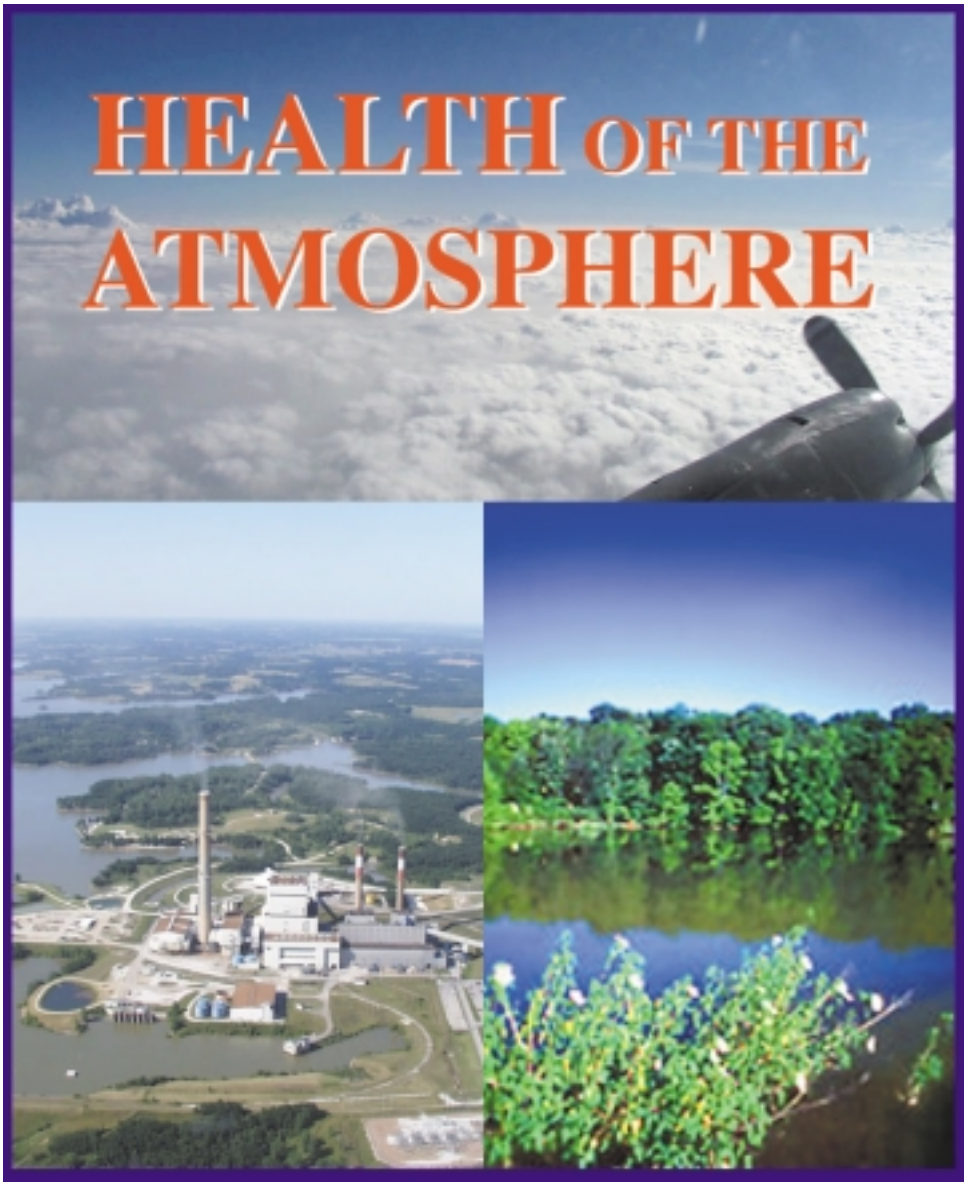

Progress Report

The First Five Years 1995-1999

HEALTH OF THE ATMOSPHERE
Progress Report – The First Five Years 1995-1999

November 1999

Copies of this report are available from:
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NOAA
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HEALTH OF THE ATMOSPHERE
Progress Report – The First Five Years 1995-1999

Aeronomy Laboratory
Air Resources Laboratory
Climate Monitoring and Diagnostics Laboratory
Environmental Technology Laboratory

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HEALTH OF THE ATMOSPHERE - AT A GLANCE

NOAA's Health of the Atmosphere research is focused on the atmospheric science that underlies regional and continental air quality with the goal of improving our ability to predict and monitor future changes, leading to improved scientific input to decision-making.

Research Goals

Characterize regional ozone episodes: Characterize the origin of ozone in rural areas, where crop and forest damage are of increasing concern [e.g., Nashville/Middle Tennessee Ozone Study, 1995; plans include future studies in the Southeast, Southwest, and Mountain West]
Document trends in air quality: Help evaluate predicted atmospheric responses to changes in emissions [i.e., the Atmospheric Integrated Research Monitoring Network (AIRMoN) and the ozone profiling network.
Develop a better understanding of the fundamental science underlying the processes responsible for the formation and distribution of fine particles in the atmosphere: Improve the atmospheric predictive capability that links sources of fine particles and their precursors to human exposure and visibility impairment.

The Participants

The foundation of the Health of the Atmosphere research program is a NOAA inter-laboratory collaboration in creative partnership with the university research community.

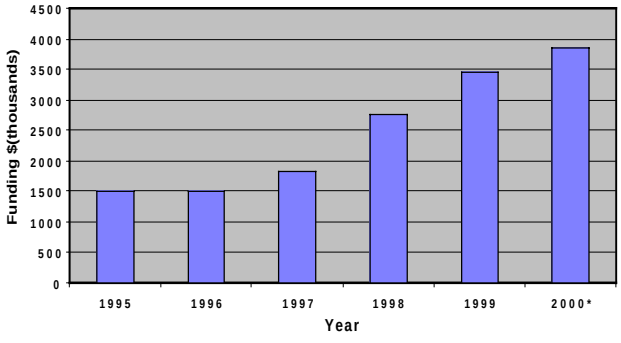
NOAA Laboratories: Aeronomy Laboratory
Air Resources Laboratory
Climate Monitoring and Diagnostics Laboratory
Environmental Technology Laboratory

Universities: University of Colorado
(Cooperative Institute for Research in Environmental Sciences)
Extramural Researchers (multiple universities)

Budget Initiative

Support for the Health of the Atmosphere research program is part of the "Long-Term Climate and Air Quality Research" line item of the NOAA budget for the Office of Oceanic and Atmospheric Research

* President's Budget



Selected Accomplishments

NOAA's research on regional chemistry and meteorology has found that…

…**natural emissions of volatile organic compounds (VOCs) from vegetation are <u>much larger</u> than human-produced emissions** of these compounds in almost all areas of the southeastern U.S. studied by NOAA. Thus, strategies focused on managing ozone through reductions in VOC emissions may not be fully effective in rural areas or in cities with abundant vegetation, such as Atlanta.

…**the contribution of natural VOCs to ozone formation was <u>significantly different</u> in different parts of the country.** The contribution was greatest in the heavily forested areas of the Southeast and less important in the farmlands of the Midwest. The mix of natural and human-influenced emissions may require consideration of different ozone management strategies in different parts of the country.

…**the previous scientific picture of the <u>chemistry of power plant plumes</u> may not be accurate**. NOAA's Health of the Atmosphere research is leading the way to a new scientific understanding of how power plant emissions effect ozone pollution. NOAA's findings are particularly timely since current ozone strategies focus on reducing emissions from rural power plants.

…**the <u>redistribution of ozone pollution</u> that occurs at <u>night</u> plays a major role in transporting urban ozone into rural areas.** Our current picture of ozone transport is dominated by what happens during the day. New observing systems developed by NOAA are changing the way we look at ozone transport; how cities effect air quality in rural areas and vice versa.

NOAA has developed an <u>atmospheric model</u> that can be used to estimate the amounts of gaseous and particulate <u>air pollution</u> that are <u>deposited</u> to sensitive ecosystems. The "Dry Deposition Inferential Method" has been adopted by the EPA for use in their Clean Air Status and Trends Network (CASTNet) and is used extensively worldwide to quantify trends in air pollution deposition.

Future Research Foci

Future research will focus on:

<u>Connections between ozone and fine particle formation</u>*:* The two most difficult air pollutants to manage are often formed and transported together. A better understanding of their interaction is critical to improved prediction.

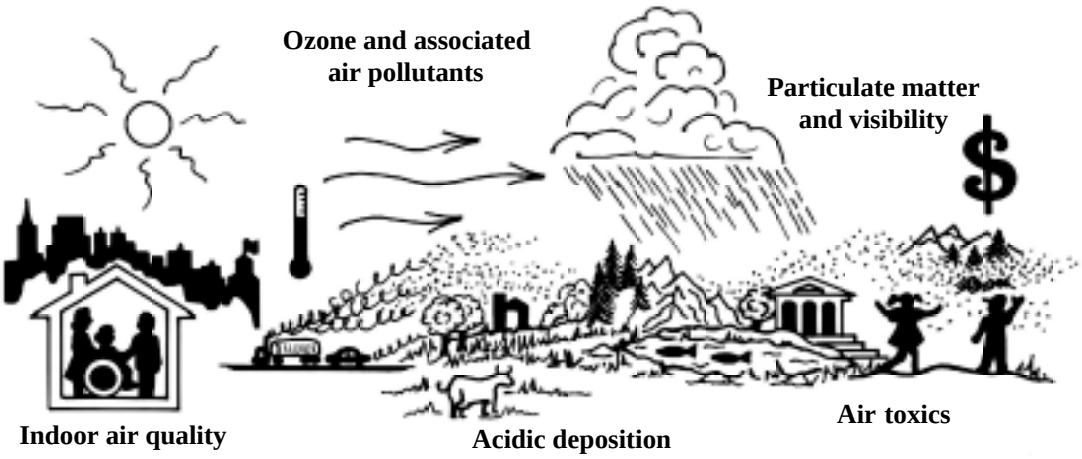
<u>Nighttime chemistry and transport</u>*:* Air pollutants can be transported long distances overnight and can "jump start" pollution episodes hundreds of kilometers downwind.

<u>Pollutant mixing and transport</u>: Current regional air quality models are limited in their ability to properly simulate pollutant mixing and transport, resulting in impaired prediction and environmental policies.

INTRODUCTION

Background

Air quality in the United States has improved dramatically in almost all respects during the three decades since the Clean Air Act was passed. Much of the oppressive pollution that characterized the Nation's cites is a distant memory. The Nation now enjoys better air quality than at any time in recent memory. However, there are still areas of the country where the Public is exposed to unhealthy levels of air pollutants and damage to sensitive ecosystems is occurring. The Air Quality Research Subcommittee of the White House's Committee on the Environment and Natural Resources (CENR) has identified[1] the five most pressing air quality issues that face the Nation today. It is in these areas that new and innovative approaches are most urgently needed if we are to manage air quality while sustaining economic growth.



Most pressing U.S. air quality issues identified by the Air Quality Research Subcommittee of the CENR[1].

While the Health of the Atmosphere research will provide new science that will contribute to a better understanding of all of these issues, the program specifically focuses on these three:

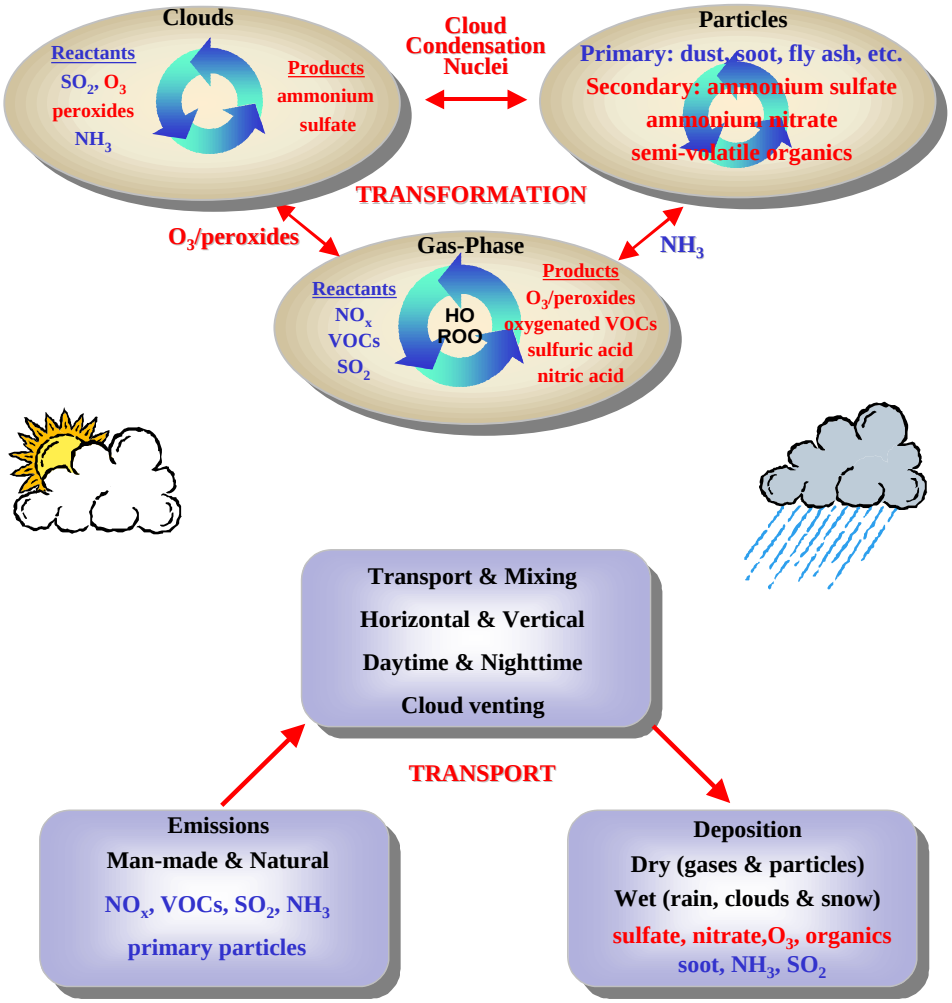
- Ozone and associated pollutants
- Particles and visibility
- Acidic deposition

Clearly, these issues are not completely independent and many of the associated phenomena are interrelated. For example, the same complex chemistry that produces

[1] Air Quality Research Subcommittee Strategic Plan, Committee on Environment and Natural Resources, National Science and Technology Council, Washington, D.C., November 1998.

ozone also results in the formation of fine particles and the acids found in acidic deposition (acid rain).

The key atmospheric processes that control the formation and distribution of pollutants in the atmosphere can be divided into two important categories, those associated with chemical and physical transformations and those associated with the transport and mixing of air pollutants. The former controls the form and phase (gas, particle, cloud) of the pollution; the latter controls the distribution (in space and time) and deposition of pollution. The ambient concentration and composition (in the case of particles, and cloud and rain drops) is a function of the complex interplay depicted below.



Atmospheric processes that control the formation and distribution of pollution in the atmosphere

Once emitted into the atmosphere, primary pollutants (those in blue above) such as sulfur dioxide (SO_2) and nitrogen oxides (NO_x) are converted in the presence of sunlight into secondary pollutants (those in red above) such as sulfuric acid, nitric acid and ozone. All

the while, this complex mix of pollutants is being transported by the winds and mixed throughout the atmosphere.

The ability to accurately forecast changes in air quality and pollution deposition that result from changes in primary pollution emissions is essential to the development of sound environmental policies. It is only through an understanding of the science that underlies these processes that such a predictive capability can be developed.

Goals and Objectives

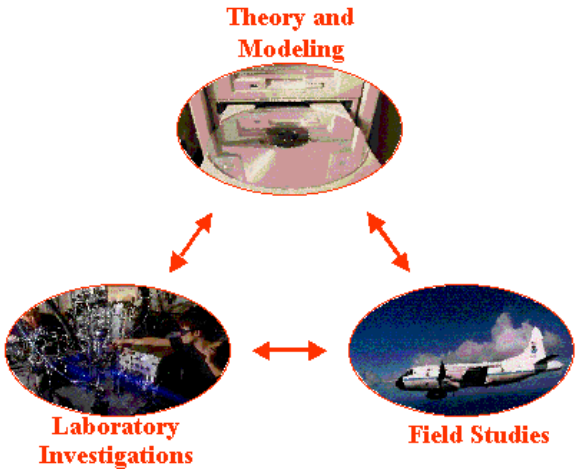
NOAA's Health of the Atmosphere research is focused on the atmospheric science that underlies regional and continental air quality, with the goal of improving our ability to predict and monitor future changes, leading to improved scientific input to decision-making.

There are three major research goals:

- Characterize regional ozone episodes: Investigate the origin of ozone in rural areas, where crop and forest damage are of increasing concern. Quantify rural/urban ozone exchange. Ozone transported from surrounding rural areas provides a "starting point" for urban ozone episodes, often contributing the majority of the measured ozone in urban areas. Conversely, ozone produced in urban areas is a major contributor to rural ozone levels.
- Determine trends in regional air quality and deposition to sensitive ecosystems and their relationship to emission reductions instituted under the Clean Air Act.
- Provide new science related to ozone and particulate matter formation and distribution: The recently enacted National Ambient Air Quality Standards (NAAQS) for ozone and particulate matter are significantly more challenging, with compliance targets set at much lower levels, and are expected to result in a significant increase in the number of areas of the country designated as "nonattainment".

Approach

The science of the Health of the Atmosphere research program is an integration of laboratory, field, and theoretical studies, a synergism that optimizes the research in each area while maximizing the overall benefits of the program. Laboratory investigations characterize and quantify fundamental properties of chemical reactions, which are needed by predictive models and point the way to specific field study approaches. Field campaigns provide the observations to test the predictive capabilities of models, as well



as indicate the potential for hitherto unknown processes that should be examined in the laboratory. Theoretical models embody the current understanding of the underlying science and, via sensitivity studies, can evaluate the impact of specific processes and emissions on continental and regional air quality. These analyses can be used to identify those areas where improved understanding will result in the biggest payoff, providing direction for laboratory investigations and aiding in the design of regional field campaigns.

The development of improved observational tools and techniques is an important part of Health of the Atmosphere research. Often, advances in understanding of these complex systems are limited by our ability to reliably characterize key atmospheric parameters. The development of new observing systems is guided by needs identified in the laboratory, field and theoretical studies described above.

The Health of the Atmosphere research is truly a team effort with contributions from the following NOAA laboratories:

- Aeronomy Laboratory – lead
- Air Resources Laboratory
- Climate Monitoring and Diagnostics Laboratory
- Environmental Technology Laboratory

The capabilities of the scientists from these laboratories are augmented by colleagues from the University of Colorado Cooperative Institute for Research in Environmental Sciences (CIRES) and a variety of other university scientists. The combination is a diverse and capable team with the technical depth and breadth and facilities needed to address the complex scientific problems presented.

Our Products

The principal "product" of the Health of the Atmosphere research program is scientific information on atmospheric processes that is needed for an improved predictive understanding of the chemical and dynamical behavior of the atmosphere. This new information is communicated to our "customers" through several mechanisms, including:

Technical presentations and peer-reviewed publications: The scientific papers that result from Health of the Atmosphere research are one of the most tangible records of our products. The listing of 1995-1998 peer-reviewed publications in Appendix A is an indication of the quality, quantity, and breadth of research that the Health of the Atmosphere investments have produced.

Web-based data sets and other data archives: The data generated in this program are a valuable resource with applications beyond those envisioned by the participating scientists. For example, data from the major field campaigns are being used by state and federal regulators in the development of local and regional management plans and by other parts of NOAA to provide improved weather forecasts.

State-of science assessments: State-of-science assessments are the primary mechanism for communicating scientific information to policymakers. The Health of the Atmosphere research strives to be "policy relevant" without being "policy driven". Therefore, we are responsible for ensuring that the results of our research are provided to decisionmakers in a "user-friendly" format. This is accomplished primarily through contributions to state-of-science assessments.

In 1996, the NARSTO (formerly known as the North American Research Strategy for Tropospheric Ozone) community committed to the preparation of a state-of-science assessment of rural ozone chemistry. This assessment is intended to review and synthesize the policy-relevant research on rural ozone, providing a consensus interpretation of the relevant science that can form the basis of effective ozone management strategies. Dan Albritton (NOAA, AL) is a member of the Synthesis Team for the assessment. Ken Schere (NOAA, ARL), the Assessment co-chair, and the following chapter authors are being supported through Health of the Atmosphere:

Daniel Jacob (Harvard University) - Heterogeneous Chemistry and Tropospheric Ozone
W.D. Neff (NOAA, ETL) - Advances in Meteorological Measurement Methods for Application to Air Quality Research and Monitoring
D.D. Parrish, F.C. Fehsenfeld (both NOAA, AL) - Methods for Gas-Phase Measurements of Ozone, Ozone Precursors and Aerosol Precursors
M. Trainer, D.D. Parrish, P.D. Goldan, J. Roberts, F.C. Fehsenfeld (all NOAA, AL) - Regional Factors Influencing Ozone Concentrations
M.L. Wesely, **B.B. Hicks** (NOAA, ARL) - Recent Progress in Dry Deposition Studies

Our Customers

The information products described above serve as input to the goals, services, missions, etc. of a variety of institutions and organizations. A few examples are provided here.

Our Colleagues: Our science is not performed in a vacuum; we benefit from those who have come before us and we contribute to a larger body of knowledge where broader perspectives and insights come from collaborative analyses. It is therefore important that we communicate the results of our research to the research communities in which we work. This is accomplished primarily through specialized scientific meetings, collaborative endeavors such as NARSTO and the Southern Oxidants Study (SOS), and peer-reviewed publication.

Governments: Air quality issues are at the forefront of the Nation's environmental agenda. The EPA has sponsored a series of regulatory initiatives that serve to "raise the bar" in terms of clean air standards. Current regulatory efforts are focused on developing management strategies for several secondary pollutants:

- *Acidic deposition*: The Clean Air Act Amendments of 1990 included provisions for the annual reduction of sulfur dioxide and nitrogen oxide emissions by 10 million and

5 million tons, respectively. Reductions are to be phased in over a ten-year period beginning in 1990.

- *Surface ozone*: In 1997, the EPA revised the air quality standard for surface ozone. The level of the standard was lowered from 120 ppb, 1-hr average, to 80 ppb, 8-hr average. This change is predicted to result in an increase in the number of areas out of compliance from 36 to 280.

- *Particulate matter*: In 1997, the EPA revised the air quality standard for particulate matter (PM) by adding a new standard that targets smaller particles ($PM_{2.5}$ – particles with aerodynamic diameters £ 2.5 microns). The new 24-hr $PM_{2.5}$ standard is 65 micrograms per cubic meter ($\mu g/m^3$) while the new annual standard is 15 $\mu g/m^3$. This change is predicted to result in a significant increase in the number of areas out of compliance.

- *Regional haze*: In 1999, EPA promulgated new regulations to improve visibility in National Parks and Wilderness Areas in the U.S. These regulations target the same fine particles that are regulated under the PM standards and will require significant reduction in fine particle levels in these pristine areas.

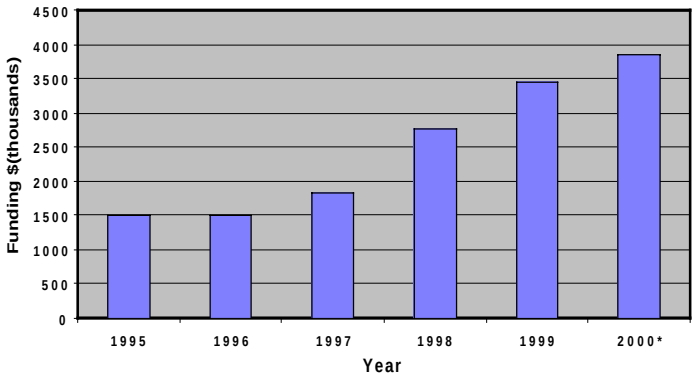
It is clear that EPA, in an effort to protect public health and the environment, is regulating air pollutants such as ozone and particulate matter at ever lower levels. As regulated levels are decreased, the roles of regional and global processes become increasingly important. Under these circumstances, NOAA's regional and continental approach to air quality, manifested through the Health of the Atmosphere research, will play a greater role by providing information that will aid EPA in the development and evaluation of regulations and assist the States in designing effective compliance strategies.

Industry: In addition to the state-of-science assessments noted above, the Health of the Atmosphere research results are an important input to industry. A notable example is the new insights on the role of rural power plants in the formation of ozone that are being developed that challenge the previous picture of plume chemistry. These findings are particularly timely since current ozone strategies focus on reducing emissions from rural power plants. NOAA is working closely with the Electric Power Research Institute and individual utilities to provide an early "heads-up", thus avoiding costly missteps.

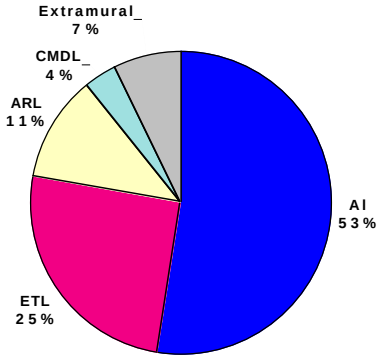
The public: We are committed to translating the results of our research into "user friendly" terms, making the science accessible to as broad an audience as possible. Our approach is to prepare short, readable publications that describe the science in lay terms and to continue to work with the media to publicize the work we do and explain its importance and implications. We also take every opportunity to describe the understanding of environmental science in schools, town meetings, and local clubs.

Funding

Since its inception in 1995, the Health of the Atmosphere program has enjoyed strong support, both from NOAA management and the Congress. In providing this support the Congress recognizes the difficult issues that face the Nation as we work to manage continental and regional air quality and the important contribution NOAA has to make in crafting workable solutions. Funding for the Health of the Atmosphere research has grown steadily over the life of the program as the value of the results has became apparent and the scope of the program has been expanded.



*President's budget



As noted above, the program is a partnership between several NOAA laboratories (led by the Aeronomy Laboratory in Boulder, CO) and the university community. The allocation of funding for FY 1999 is shown on the left. Funding for CIRES scientists is included in the budgets for the NOAA laboratories.

A NOAA WP-3 "hurricane hunter", doing double duty as a flying air quality laboratory, works in the haze over Tennessee during the 1995 Southern Oxidants Study field campaign. A large coal-burning power plant can be seen in the background. The unique capabilities of this aircraft and its extensive array of on-board instrumentation has led to important new insights into the atmospheric processes that influence regional air quality.

SELECTED ACCOMPLISHMENTS

Testing the importance of natural emissions from forests on regional ozone formation

The relative importance of natural volatile organic compounds (VOCs). If ozone (O_3) concentrations are to be successfully managed by the implementation of primary emission controls, it is essential that the role of naturally produced VOCs vis-a-vis the manmade VOCs be thoroughly understood. Hardwood forests, particularly those in the South, emit large quantities of isoprene, a very reactive VOC that readily produces ozone when mixed with nitrogen oxides (NO_x) in the presence of sunlight. It has become clear in the past decade through our research that while most of the NO_x involved in regional pollution is anthropogenic in origin, both biogenic or natural (BHC) and anthropogenic or man-made (AHC) hydrocarbons contribute to regional ozone formation. The relative importance of BHCs and AHCs to O_3 formation must be properly understood so that effective control strategies for regional ozone can be formulated.

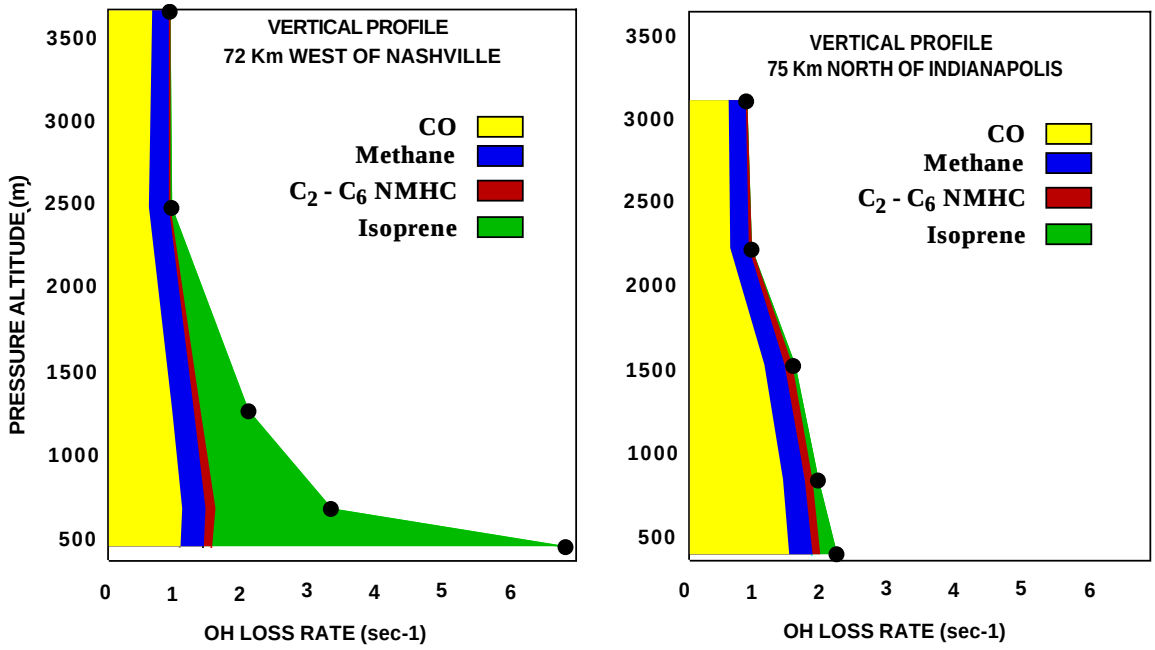
Measurements of products provide an indicator. Our research has provided a method to address this problem by providing an observational means to estimate the impact that BHCs and AHCs have on O_3 formation chemistry. This method relies on measurements of three related peroxy nitrate compounds, PAN, PPN, and MPAN, that are bi-products of the same chemistry that produces ozone. However, MPAN is a unique product of isoprene, the most important reactive BHC, while PPN is essentially solely produced from AHCs and PAN is a general product of both BHC and AHC chemistry. The simultaneous measurement of these product species provides the opportunity to determine the relative effectiveness of biogenic as opposed to anthropogenic compounds in ozone-forming atmospheric chemistry. The three compounds were measured aboard the NOAA WP-3 aircraft during the 1995 Southern Oxidants Study in and around Nashville, Tennessee, along with O_3, NO_x, hydrocarbons, and a number of other chemical and meteorological quantities. These represent the first airborne measurements of PAN, PPN and MPAN. Moreover, this capability was used to arrive at quantitative estimates of the effect of BHC and AHC chemistry on O_3 formation.

What was found? From these measurements, it was found that isoprene chemistry dominated ozone formation in the forested rural areas of the Nashville/Middle Tennessee region. However, the contribution of isoprene to rural ozone formation was found to decrease significantly with increasing latitude (i.e., from south to north). This decrease paralleled a similar decrease in isoprene emissions with latitude as predicted by current biogenic emission inventories derived from inventories of forest density and type.

Williams, J., J.M. Roberts, F.C. Fehsenfeld, S.B. Bertman, M.P. Buhr, P.D. Goldan, G. Hübler, W.C. Kuster, T.B. Ryerson, M. Trainer, and V. Young, Regional ozone from biogenic hydrocarbons deduced from airborne measurements of PAN, PPN, and MPAN, *Geophysical Research Letters*, **24**, 1099-1102, 1997.

Role of carbon monoxide and methane in regional ozone formation

Long range transport of longer-lived ozone precursors can be important. It is well known that the elevated ozone concentrations that frequently are found during the summer over the eastern United States are potentially harmful to humans and damaging to crops and forests. It is now believed that during the summer most ozone in the eastern U.S. is produced photochemically by the interaction of sunlight on ozone precursors. However, once produced, the lifetimes of ozone and certain ozone precursors are sufficiently long so that they can be transported over great distances. For this reason, ozone concentrations in a particular location may depend on the amount of ozone and ozone precursors transported from outside the local region.



Importance of carbon monoxide relative to non-methane hydrocarbons (NMHCs) as an ozone precursor evaluated over a wide region. An analysis of the data taken during the course of regional flights over the southeastern and midwestern U.S. during the 1995 Nashville/Middle Tennessee Study of the Southern Oxidants Study suggests that carbon monoxide makes a significant contribution to ozone formation in regions where isoprene levels are depressed (i.e., in the boundary layer in urban plumes, in the upper Midwest and in the free troposphere). The more reactive VOCs have high removal rates and their absence suggests a lack of nearby sources of these VOCs. Both CO and CH_4 have a variety of natural and anthropogenic sources and are produced by the oxidation of more complicated hydrocarbons. Moreover, during a typical midsummer period, both CO and CH_4 are long-lived compounds with atmospheric lifetimes of one to several months. Hence, the exact sources of these compounds at a particular location will be difficult to determine.

Carbon monoxide can contribute significantly to photochemical ozone formation. The apparent importance of methane and CO in ozone formation in the regions described above necessitates their consideration in future ozone management strategies. The role of CH_4 and CO should be expected to be further enhanced as emissions of more reactive VOCs continue to decline.

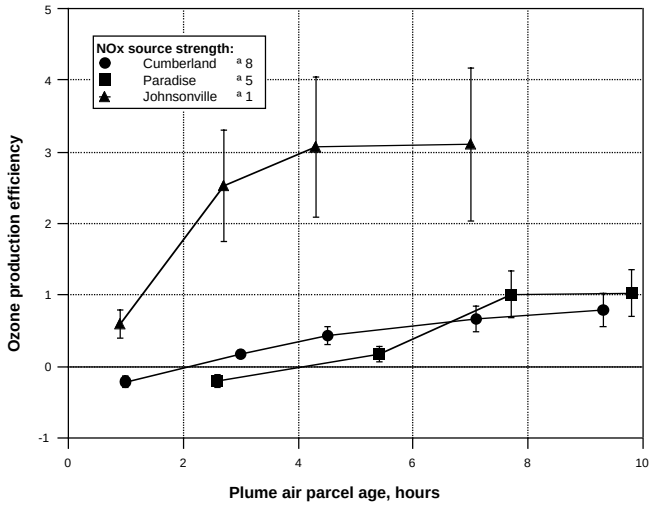
Golden, P.D., D.D. Parrish, W. Kuster, M. Trainer, S. McKeen, J. Holloway, T. Jobson, D. Sueper, and F.C. Fehsenfeld, Airborne measurements of isoprene, CO, and anthropogenic hydrocarbons and their precursors, *Journal of Geophysical Research*, in press, 1999.

Trainer, M., D.D. Parrish, P.D. Goldan, J. Roberts, and F.C. Fehsenfeld; Regional factors influencing ozone concentrations, *Atmospheric Environment*, in press, 1999.

Inefficient ozone formation in plumes

NO_x reduction to control ozone. The photochemical production of ozone requires nitrogen oxides (NO_x). Hence, a control strategy that is presently being pursued is to reduce the photochemically formed ozone by reducing the emission of NO_x. This is being done by reducing the emissions of NO_x from large point sources such as power plants, which on a nationwide basis contribute a substantial fraction of the NO_x emitted from all sources. Since O_3 production is non-linear with the amount of NO_x emitted, the wisdom of this approach requires further scrutiny.

Results from studies of power plant plumes. These measurements were made as part of the 1995 Nashville/Middle Tennessee Study of the Southern Oxidants Study (SOS). Cross-plume pollutant profiles obtained by the NOAA WP-3D were combined with detailed wind fields to investigate O_3 formation at various downwind distances for several pollutant sources with very different NO_x emissions. The analysis of these data from the measurements that were made in 1994 and 1995 indicate that NO_x removal occurred quickly and that O_3 production in these plumes was much less efficient than ozone production from more dispersed NO_x sources. Ozone production per unit of NO_x emission appears to be greatest for the Nashville urban plume and for the smaller NO_x emitting power plants that are located in areas rich in natural VOC emissions. Ozone production was found to be less efficient for NO_x emitted from rural power plants with the higher NO_x emissions.



Ozone production efficiency as a function of NO_x emission rate for three power plants.

Implication for emission controls. The implications of these findings are significant in terms of strategies that are being proposed to manage ozone by regulating sources of NO_x. The approach currently being pursued would reduce NO_x emissions by reducing the emission of NO_x from large point sources. However, our results suggest that the effectiveness in reducing photochemically produced ozone pollution may be substantially less than what might be expected with relatively large reductions in NO_x. These data suggest that one could get more "bang for the buck" by controlling smaller sources. The results also suggest that the trading of NO_x emissions from power plants over long distances may be of limited value in reducing ozone nonattainment, since ozone production appears to occur relatively near the source.

Ryerson, T.B., M.P. Buhr, G. Frost, P.D. Goldan, J.S. Holloway, G. Hübler, B.T. Jobson, W.C. Kuster, S.A. McKeen, D.D. Parrish, J.M. Roberts, D.T. Sueper, M. Trainer, J. Williams, and F.C. Fehsenfeld, Emissions lifetimes and ozone formation in power plant plumes, *Journal of Geophysical Research*, **103**, 22569-22584, 1998.

Testing the mechanisms for isoprene oxidation

The importance of isoprene oxidation. The regional ozone pollution problem has long resisted control and management strategies and remains one of our most important air quality issues. Reasons for the persistence of this problem have to do with the fact that O_3 is not emitted directly from pollution sources, but is produced photochemically from the oxides of nitrogen (NO_x) and VOCs that are derived from a variety of sources. During the last decade, largely as a result of NOAA research, there has been a growing appreciation for the role of summertime biogenic VOC emissions from vegetation in photochemical ozone formation, both in heavily forested urban areas as well as in rural areas. These biogenic VOC emissions are largely in the form of isoprene, a highly reactive, unsaturated hydrocarbon. Our studies indicate that the influence of the oxidation of this isoprene on regional ozone formation can be great. Clearly, therefore, it is essential that the mechanisms developed through laboratory kinetic measurements that

are thought to control isoprene oxidation be subjected to independent careful verification by field measurements.

Observations of the oxidation products confirm oxidation mechanisms. Laboratory measurements have indicated that methacrolein, methyl vinyl ketone and 3-methyl furan are the principal molecular byproducts directly formed by the HO-initiated oxidation of isoprene. Measurements of these compounds along with isoprene were carried out at a rural site in Alabama. The analysis of these measurements, using a one-dimensional model simulation, showed that their relative and absolute concentrations are in good agreement with the present understanding of the photochemistry of isoprene. The model interpretations of these measurements also indicated that the oxidation of isoprene could adequately account for all the methacrolein, methyl vinyl ketone and 3-methyl furan that was observed.

Implication of the research. In addition to the measurements of the specific peroxy-nitrate compound MPAN that is formed in the oxidation of methacrolein, these measurements confirm the oxidation schemes for isoprene that have been developed in the laboratory and support the importance of the role that isoprene can play in the regional formation of ozone. It should also be noted that the development of effective ozone control strategies is dependent on the knowledge of the emission rate of natural hydrocarbons such as isoprene in urban as well as rural areas. Due to its high photochemical reactivity, the concentrations of isoprene are spatially highly variable and are influenced by vegetation in close vicinity of the measurement location. However, due to their longer photochemical lifetime, the concentrations of methacrolein and methyl vinyl ketone will be representative for the emissions of isoprene over a larger fetch. Ambient measurements of these oxidation products in settings with highly inhomogeneous distribution of isoprene-emitting vegetation, such as urban areas, provide a more stringent test of the role that isoprene plays in urban and regional ozone formation.

Montzka, S.A., M. Trainer, W.M. Angevine, and F.C. Fehsenfeld, Measurements of 3-methyl furan, methyl vinyl ketone, and methacrolein at a rural forested site in the southeastern United States, *Journal of Geophysical Research*, **100**, 11393 - 11401, 1995.

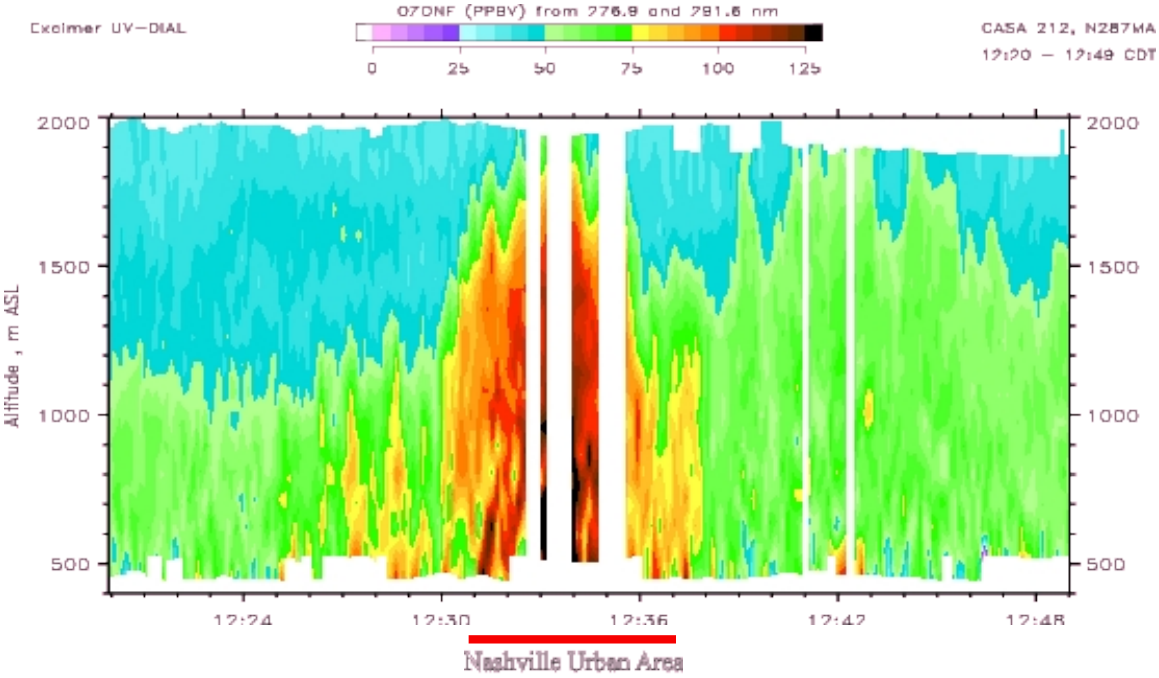
Frost, G.J., M. Trainer, G. Allwine, M.P. Buhr, J.G. Calvert, C.A. Cantrell, F.C. Fehsenfeld, P.D. Goldan, J. Herwehe, G.F. Hübler, W.C. Kuster, R. Martin, R.T. McMillen, S.A. Montzka, R.B. Norton, D.D. Parrish, B.A. Ridley, R.E. Shetter, J.G. Walega, B.A. Watkins, H.H. Westberg, and E.J. Williams, Photochemical Ozone production in the rural southeastern United States during the 1990 ROSE program, *Journal of Geophysical Research*, **103**, 22491-22508, 1998.

Ozone and fine particle formation and distribution during stagnation episodes

Urban pollution accumulates during stagnations. Conditions that lead to the highest hourly urban ozone concentrations are characterized by abundant sunshine and light

winds. Under these conditions, pollutants accumulate near the source region and can reach levels in excess of Federal standards. The very highest concentrations are expected during the lightest winds, i.e., during stagnation conditions. During the 1995 SOS Nashville campaign, a 3-day stagnation episode occurred in mid July. NOAA scientists employed an airborne O_3 differential-absorption lidar (DIAL) to study the formation and transport of ozone and aerosols under these conditions at a level of detail that has not previously been possible.

The airborne lidar provides unique insights into pollution formation and distribution. The lidar provides vertical cross sections of O_3 concentration and aerosol backscatter along the flight path of the aircraft. Examples of data collected during the July 1995 stagnation episode are provided below.



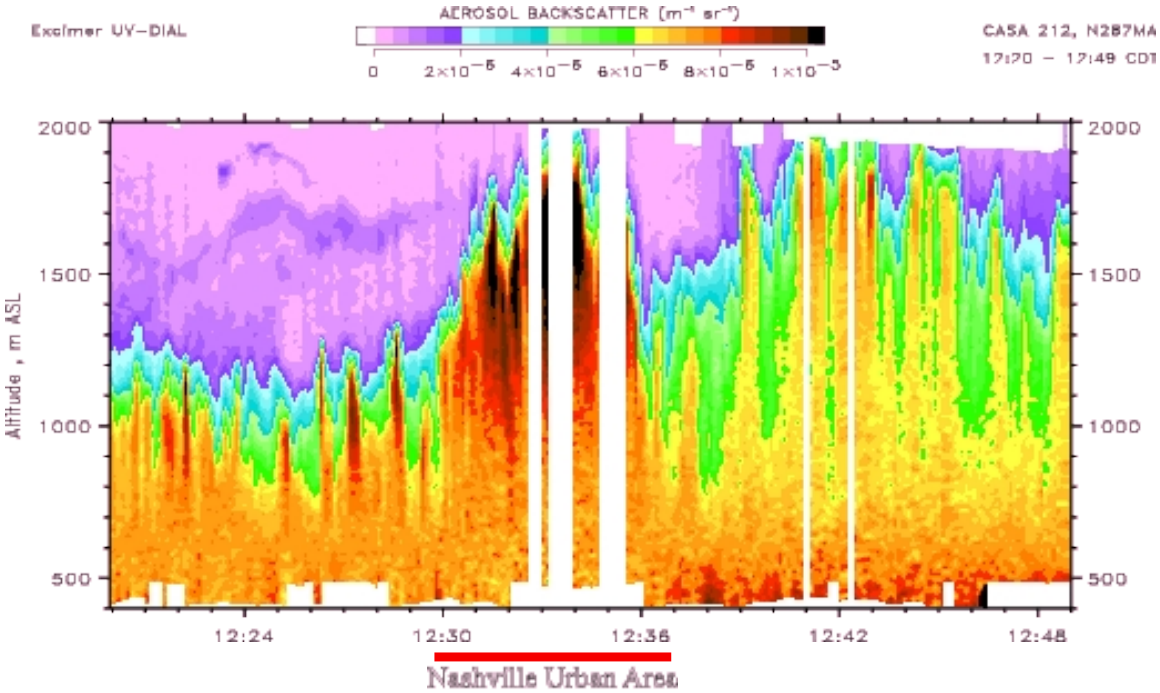
Vertical cross section of O_3 concentrations (ppbv) from NW to SE over Nashville on 12 July 1995. Abscissa is in hours (CDT) and minutes of flight time; each minute represents 4 km distance, so entire leg is ~110 km.

What have we learned? Analysis of O_3 and aerosol backscatter cross sections obtained during the stagnation has resulted in a clearer view of the processes responsible for pollution accumulation.

- *Daytime accumulation.* Cross sections over the city on 12 July, the middle day of the 3- day episode, showed that the urban pollutants formed a dome over the urban area. Significant O_3 reached ~1.6 km above the surface, and O_3 concentrations exceeding 120 ppb extended to >1.2 km. The presence of this deep mixed layer mitigated the ozone concentrations, to some extent, by providing a larger mixing volume. A horizontal plot of O_3 along the flight track shows that the pollution remained very

close to Nashville. The data clearly indicate that, at least on this occasion, emissions from the Nashville urban area were responsible for most of the ozone over the city.

- *Variability in mixing height.* Another important result of the light horizontal winds during stagnation is that local differences in land use can express themselves as local differences in mixing height. On the middle day of the episode a difference of ~500m in mixing height was noted over a horizontal distance of a few 10's of kms between the forested land to the west through northwest of Nashville and the farming and pasture land in the other directions around the city. This observation has significant implications for both observed pollution levels and transport across these regions of varied land use.



Aerosol backscatter cross section from flight leg shown above. Mixing height *h* is height where larger values in mixed layer decrease rapidly with altitude to smaller values aloft. Cross section shows lower *h* values to NW of city (left part of figure) than over or to the SE of the urban area.

Banta, R.M., C.J. Senff, A.B. White, M. Trainer, R.T. McNider, R.J. Valente, S.D. Mayor, R.J. Alvarez II, R.M. Hardesty, D.D. Parrish, and F.C. Fehsenfeld, Daytime buildup and nighttime transport of urban ozone in the boundary layer during a stagnation episode, *Journal of Geophysical Research*, **103**, 22519-22544, 1998.

The production, destruction, and transport of ozone in power plant plumes

Rural power plants produce a significant fraction of the Nation's NO_x emissions. EPA's new "NO_x rule" targets fossil-fueled power plants in the East in an effort to reduce regional ozone levels. A better understanding of the contribution of these sources to regional pollution levels will aid in an evaluation of this approach.

Which measurements were performed to characterize power plant plumes? The NOAA/ETL's airborne O_3 and aerosol lidar was used in conjunction with *in situ* sampling to investigate power plant plumes in the Nashville area during the 1995 SOS campaign. The downward-looking lidar was used to map out the two-dimensional structure of the plume at multiple distances downwind from the plant, providing information on plume size, shape, and the vertical distribution of ozone and aerosols. The plume location with respect to the top of the boundary layer was determined using the lidar aerosol returns to provide information on the boundary layer height. In addition, the lidar ozone data was used to calculate ozone production rates in the plume as a function of distance downwind.

What we learned. The location and transport of the plumes was a function of the height and strength of the capping inversion. On one occasion, when there was strong vertical mixing, the power plant plume was spread across the entire depth of the boundary layer but did not reach into the free troposphere. The plume was shaped symmetrically and had a clearly defined, single core (Fig. 1). On another occasion, part of the power plant plume had penetrated the inversion capping the boundary layer. Subject to different meteorological conditions, the two portions of the plume above and below the boundary layer top evolved differently as they were transported downwind, resulting in a very irregular shape and a near breakup of the plume (Fig. 2). In both cases, we found that close to the power plant O_3 was destroyed in the plume, while farther downwind ozone was produced at rates of up to 4 ppb/hr. The latter confirms that NO_x-rich power plant plumes have the potential of raising local ozone levels significantly over the course of a day.

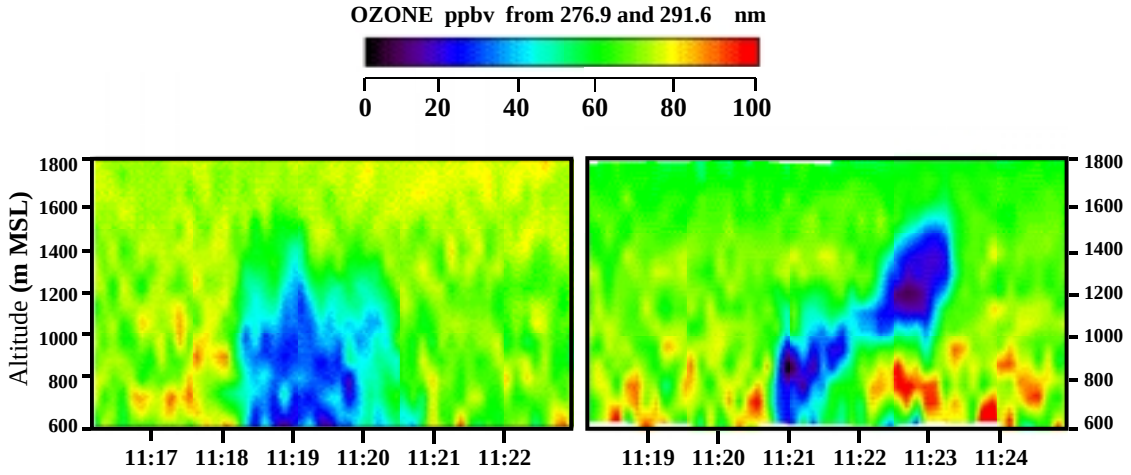


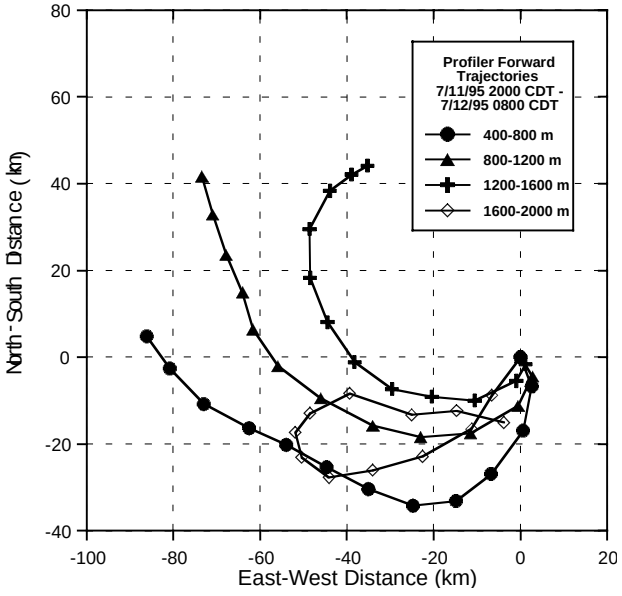
Fig. 1 **Fig. 2**

Senff, C.J., R.M. Hardesty, R.J. Alvarez II, and S.D. Mayor, Airborne lidar characterization of power plant plumes during the 1995 Southern Oxidants Study, *Journal of Geophysical Research*, in press, 1999.

Nighttime winds are a key factor in daytime ozone levels

Stagnation episode. The highest ozone concentrations measured during the 1995 SOS Nashville/Middle Tennessee Ozone Study (138 ppb – 1-hr avg.) occurred during a period of intense stagnation in mid July. However, during this period, midmorning O_3 concentrations were much lower, suggesting that pollution that had accumulated over the city the previous day had been transported away during the night.

The role of nocturnal transport. While daytime winds were weak during the stagnation episode (80% of the wind speeds measured in the boundary layer were less than 3 m s^{-1}), nighttime winds above the nocturnal boundary layer accelerated to 5–10 m s^{-1}, as they became decoupled from the surface-induced friction. In addition, the nocturnal winds rotated in time and with height in accordance with the principles of the inertial oscillation. These rotations are visible, for example, in the forward trajectories calculated from the boundary-layer wind profiler network. The pandemic nature of the inertial oscillation during the 1995 field study was demonstrated by McNider *et al.* (1998). They computed energy spectra from the velocity components measured with the wind profilers for the entire campaign. These spectra exhibited a prominent peak at a period of ~20 hr, which agrees with the expected inertial period for Nashville. The important role of these nocturnal accelerations in redistributing the Nashville urban plume was described by Banta *et al*. (1998). They used the wind profiler trajectories shown below to predict the overnight movement of the urban plume on 11 July. Enhanced ozone was detected on the morning of 12 July by airborne DIAL and *in situ* measurements near the locations and altitudes predicted by the trajectories.

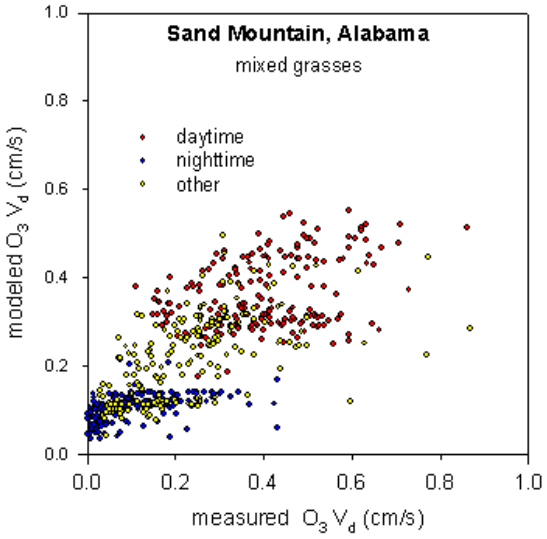


Banta, R.M., C.J. Senff, A.B. White, M. Trainer, R.T. McNider, R.J. Valente, S.D. Mayor, R.J. Alvarez II, R.M. Hardesty, D. Parrish, and F.C. Fehsenfeld, Daytime buildup and nighttime transport of urban ozone in the boundary layer during a stagnation episode, *Journal of Geophysical Research*, **103**, 22519 - 22544, 1998.

McNider, R.T., W.B. Norris, A.J. Song, R.L. Clymer, S. Gupta, R.M. Banta, R.J. Zamora, A.B. White, and M. Trainer, Meteorological conditions during the 1995 Southern Oxidants Study Nashville/Middle Tennessee field intensive, *Journal of Geophysical Research*, **103**, 22225 - 22243, 1998.

Better estimates of dry deposited nitrogen and sulfur.

The NOAA Atmospheric Integrated Research Monitoring Network (AIRMoN) provides a research-based foundation for routine atmospheric deposition monitoring operations [National Atmospheric Deposition Program (NADP) for wet and the Clean Air Status and Trends Network (CASTNet) for dry]. Although well-documented techniques exist for monitoring wet deposition, dry deposition monitoring continues to evolve since there are no exiting methodologies that are suitable for routine operation. The Dry Deposition Inferential Method (DDIM) has been and continues to be the method of choice for dry deposition monitoring, not only for North American networks but also for monitoring activities worldwide. With this method, dry deposition rates of gas and aerosol phases of sulfur, nitrogen, and ozone are derived by coupling measurements of air concentrations with a modeled deposition velocity. The deposition velocity depends on the pollutant



species, as well as on atmospheric and land surface data that are indicative of the dry deposition processes. Since implementation of the inferential method requires the use of a deposition model, considerable effort has been made to evaluate the bias and uncertainty of the model. This requires special field studies in which the dry deposition rates of key pollutants are measured directly, usually with micrometeorological methods. The deposition velocities from these three intensive field campaigns, in which the fluxes of SO_2, O_3 and HNO_3 were measured directly, recently were compared with estimates from a land/surface numerical model, denoted as the multi-layer model. An example comparison between modeled and measured deposition velocities for a grassland site is shown above. For O_3, the overall model bias was low and not significantly different from zero. The model generally underpredicts the deposition for SO_2 and HNO_3 by 15% and

20%, respectively. Planning for additional studies to examine the predictive capability of the model when applied to aerosol deposition in marine environments is currently underway.

Meyers, T.P., P. Finkelstein, J. Clarke, T.G. Ellestad, and P.F. Sims, A multi-layer model for inferring dry deposition using standard meteorological measurements, *Journal of Geophysical Research*, **103**, 22645-22661, 1998.

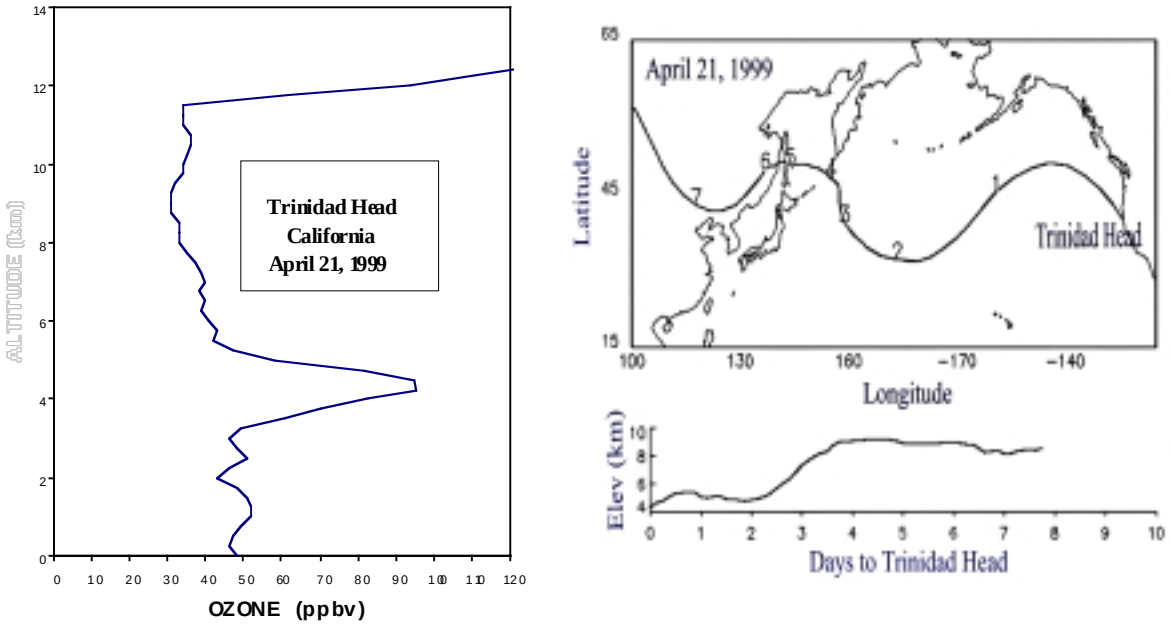
Ozone in the background troposphere over the U.S.

Changes in ozone in the troposphere over the U.S. Although the ozone concentration near the ground is widely measured across the continental U.S., only a small number of sites make regular measurements of the ozone content of the troposphere above the boundary layer. Long-range transport of ozone from distant sources as well as stratospheric input to the troposphere are processes that can only be understood with measurements in the free troposphere. At two locations, Wallops Island, Virginia (a site operated by NASA) and Boulder, Colorado (operated by NOAA) regular ozone profiles have been obtained for 15 or more years. At both of these sites there have been only small changes in the ozone content of the troposphere.

Transport of ozone across the Pacific. In August 1997 an ozone profiling station was established on the U.S west coast in northern California at Trinidad Head. This site was chosen because of its exposure to air entering the U.S. with the prevailing westerly flow. During April, May, and June there are episodes of rather direct flow from eastern Asia. Adequate sunlight for photochemical ozone production is also available. Under these conditions enhanced ozone levels may be observed entering the U.S. and can contribute to the overall tropospheric ozone background levels. An example of such an enhanced layer is seen in the profile from April 21, 1999 at Trinidad Head. The back trajectory at the level of the ozone peak shows that air parcels could have come from the Asian continent approximately 6-7 days earlier.

Tropospheric ozone in the southeastern U.S. In support of the two Nashville SOS field campaigns, near-daily ozone profiles were obtained that gave ozone amounts through both the troposphere and stratosphere. These profiles showed strong low altitude ozone enhancements associated with the pollution buildup of ozone, as well as occasional intrusions at higher tropospheric altitudes that appear to be associated with transport from the stratosphere. Ozone profiles are also being obtained regularly from a recently established site in Huntsville, Alabama.

Oltmans, S.J., A.S. Lefohn, H-E. Scheel, J.M. Harris, H. Levy II, I.E. Galbally, E.-G. Brunke, C.P. Meyer, J.A. Lathrop, B.J. Johnson, D.S. Shadwick, E. Cuevas, F.J. Schmidlin, D.W. Tarasick, H. Claude, J.B. Kerr, O. Uchino, and V. Mohnen, Trends of ozone in the troposphere, *Geophysical Research Letters*, **25**, 139-142, 1998.



Ozone profile at Trinidad Head, California on April 21, 1999, with the isentropic back trajectory ending at 4.3 km over the ozonesonde site. The numbers on the trajectory path represent the number of days back from the time of the observation.

OUTREACH

The NOAA Health of the Atmosphere research program reaches out to the larger stakeholder community through a series of collaborative partnerships. These strategic alliances provide opportunities to leverage NOAA's science to a larger community and provide important linkages to air pollution effects research and regulation and policy development. Current research collaborations include:

The Federal Sector - through the White House's Committee on Environment and Natural Resources (CENR). NOAA currently provides leadership to this committee as Co-Chair of the CENR and as Vice Chair of the CENR's Air Quality Research Subcommittee (AQRS). The Air Quality Research Subcommittee has designated PM and ozone research as the highest priority for the FY 1999 and FY 2000 budget periods.

National and International - through leadership in forming NARSTO (formerly called the North American Research Strategy for Tropospheric Ozone). This is a tri-national (Mexico, United States, and Canada) organization, a public/private partnership, whose membership spans government, the utilities, and academe. Its primary mission is to coordinate policy-relevant scientific research with a goal of determining workable, efficient, and effective strategies for local and regional ozone and PM management. NOAA is playing a leadership role in the development of an assessment of policy-relevant scientific information on ozone.

Particulate Matter Measurements Workshop - NOAA has played a leadership role in bringing together scientists from the atmospheric, exposure, and health effects communities to identify the key components and design parameters for a comprehensive measurement program to characterize ambient particulate matter and important co-pollutants. In July 1998, Dan Albritton of NOAA's Aeronomy Laboratory and Dan Greenbaum of the Health Effects Institute co-chaired a NARSTO/EPA sponsored workshop to address these issues in Chapel Hill, North Carolina. The recommendations of this workshop are reported in *Atmospheric Observations: Helping Build the Scientific Basis for Decisions Related to Airborne Particulate Matter.*

Research Partnerships: NOAA plays a key role in several research consortia that are involved in air quality research, including:

Southern Oxidants Study (SOS) - NOAA was one of the founders of the SOS program, which was initiated in 1988 to provide scientific research to develop effective ozone reduction strategies for the Southeast and the Nation. The program involves federal and state agencies, university research groups, and interested parties from the private sector. Active participation by federal, state, and local regulators in the SOS affords a direct

linkage between the scientific and policy communities, ensuring focused research and timely implementation of new insights that have been developed.

National Atmospheric Deposition Program/National Trends Network (NADP/NTN) - The NADP/NTN consists of nearly 200 sites measuring precipitation chemistry, operated by over 100 organizations, including federal, state and local agencies, universities, and industrial groups. NOAA's Atmospheric Integrated Research Monitoring Network (AIRMoN) serves as a research component of the larger NADP precipitation chemistry network, providing higher frequency measurements at a limited number of locations. These networks provide early detection of improved air quality resulting from sulfur and nitrogen emission reductions that were implemented as part of the Clean Air Act Amendments of 1990.

Clean Air Status and Trends Network (CASTNet) – The CASTNet consists of 71 monitoring stations located in rural areas to assess broad air quality trends. NOAA and the EPA are working together to integrate air quality data from AIRMoN and CASTNet to provide a more comprehensive picture of the Nation's air quality, with a focus on documenting changes associated with emission reduction programs initiated under the Clean Air Act. Both CASTNet and AIRMoN use methodologies developed and validated by NOAA to translate weekly ambient concentrations into dry deposition estimates.

Extramural Programs:

NOAA's Health of the Atmosphere program provides support to university and private-sector scientists conducting research that is closely linked to the program's objectives. The extramural research program extends NOAA's own capabilities and provides opportunities for collaboration that enrich the science of both NOAA and the performing organizations. A sampling of the university research being supported under the Health of the Atmosphere program is provided below.

University of Virginia, Keene, Moody, Cosby, and Galloway – Analyze air quality and deposition data from the AIRMoN network to determine the impact of recent emission reductions.

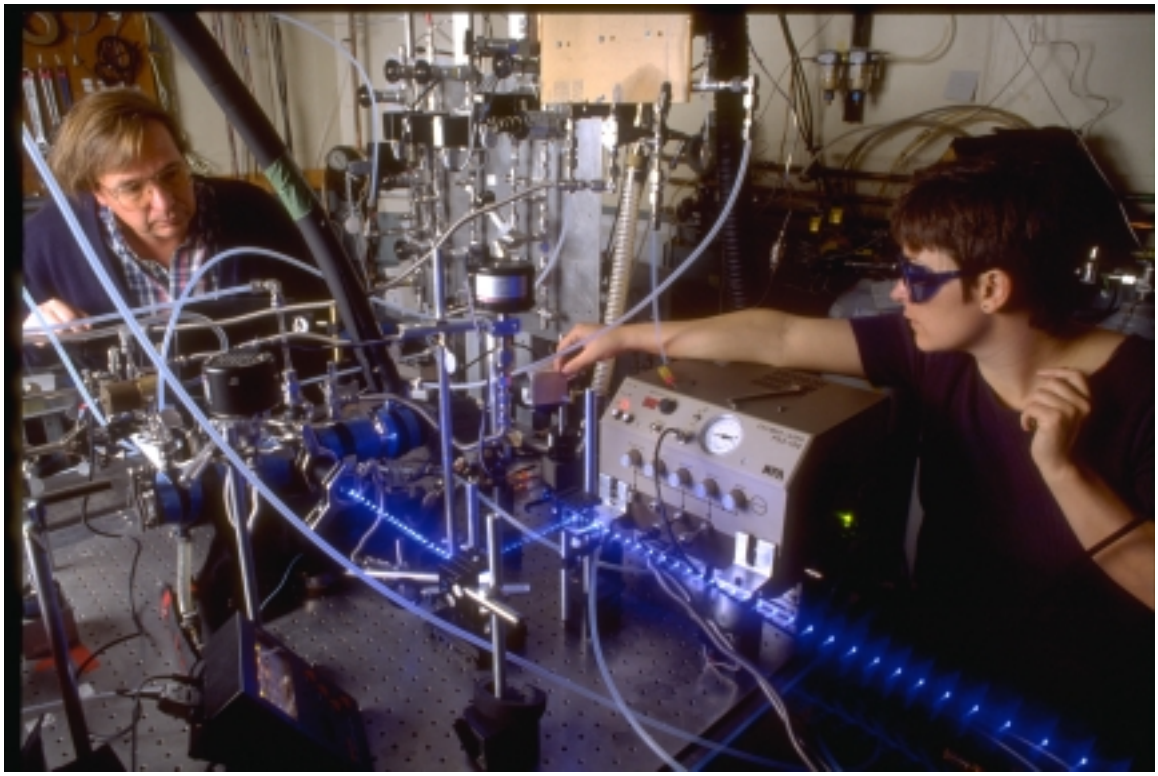
Pennsylvania State University, Brune – Measure hydroxyl and hydroperoxyl radical concentrations (key intermediates in ozone and fine particle formation) during the 1999 SOS field campaign in Nashville, Tennessee.

Purdue University, Shepson – Use measurements performed during the 199 SOS field campaign in Nashville, Tennessee to evaluate the role of naturally-occurring hydrocarbons.

Western Michigan University, Bertman – Perform measurements of organic nitrates during the SOS field campaign in Nashville, Tennessee to determine ultimate fate of NO_x emissions.

Ohio University, Young – Characterize the contribution of C_2-C_8 hydrocarbons to local and regional oxidant chemistry during the SOS field campaign in Nashville, Tennessee.

University of Alabama at Huntsville, McNider – Develop and evaluate a large eddy simulation (LES) model with coupled chemistry.



The laboratory study of chemical and physical processes that underlie the transformation of air pollutants from one form to another is an integral part of Health of the Atmosphere research. Results of these fundamental studies and those from intensive field studies and modeling studies are integrated within the program to provide new perspectives and understanding of the complex processes that control the formation and distribution of ozone and fine particles.

FUTURE PLANS

Research Objectives

Connections between ozone and PM formation: Anthropogenic pollution can be harmful to human health, have deleterious effects on crops and forests, and degrade visibility. At present, the most serious air quality concerns deal with ozone and fine particle pollution. All of the ozone and much of the aerosol that are attributed to human pollution are not emitted directly into the atmosphere but are produced as byproducts of atmospheric chemistry. The aim of the Health of the Atmosphere research over the near future is to investigate how the chemical processing and removal of compounds of anthropogenic origin can influence the regional budgets of ozone and fine particles.

The Health of the Atmosphere findings described above have made a significant impact on national policies developed to control ozone on local and regional scales in the United States. Their general applicability to all locales and regions in the U.S. requires additional testing and verification. In addition, these unexpected conclusions force us to consider other factors that may influence the photochemical processing of these compounds. In particular, the role of aerosols in tropospheric chemistry needs to be addressed.

During the next two years, the Health of the Atmosphere program will sponsor field measurements of the concentrations of O_3, fine particles, their precursors, photochemical intermediates and other photochemical products, and other atmospheric parameters. Airborne and ground-based studies will provide information required to more accurately determine the rate and amount of ozone and fine particulate matter that are produced in the atmosphere from natural and human-made precursors.

The expansion of our measurements to investigate aerosol and aerosol-related processes is timely. Aerosols participate in a variety of chemical and physical processes in the troposphere. On a regional scale, these processes are associated with regional air quality as related to visibility and the effects of fine particles on human health. In this regard, there is a natural synergism between the ozone-related research described previously and the study of processes leading to or involving fine particles. New federal standards have been proposed for both ozone and fine particles. These standards force us to recognize that a basic scientific understanding of the chemistry and physics of the atmosphere is prerequisite in order to design effective control strategies for these pollutants and that the concentrations of the pollutants in the atmosphere are often co-dependent because of interacting chemical reactions.

With this in mind, we propose to expand our measurements program to elucidate:

- how chemical processing on aerosols influences ozone formation
- how the atmospheric oxidation leading to ozone formation leads to aerosol formation

- how atmospheric chemistry influences the growth and chemical composition of aerosols.

This new paradigm also has been embraced by our colleagues in The Southern Oxidant Study and NARSTO.

This comprehensive research program will investigate the relationship between the sources of fine particles and ozone precursors and photochemistry that lead to ozone and fine particle formation. It is designed to continue and expand past investigations that were undertaken as part of NARSTO and the Southern Oxidants Study. The measurements we plan are intended to provide a better understanding of the basic chemical, meteorological, and transport processes that determine ozone and fine particle distributions and to use this information to assist policy makers in devising optimal ozone management strategies.

Nightime Chemistry and Transport: Because of the important role that photochemistry plays in the formation of ozone and other secondary air pollutants, the vast majority of the intensive atmospheric process studies conducted to date have focused on the daylight hours. Analysis of data collected during the 1995 Nashville/Middle Tennessee ozone study revealed that nighttime mixing and transport playes a significant role in the redistribution of urban pollution throughout the region. During the day, under the stagnant conditions associated with most pollution episodes, only limited horizontal transport of the pollution plumes is expected. However, at sunset a shallow nocturnal inversion layer may form that can isolate the boundary layer pollution from the surface, reducing the loss of ozone, PM and their precursors via deposition. The formation of this layer also reduces the frictional resistance for the air masses above the nocturnal layer, allowing this pollution to be more effectively transported over larger distances. Moreover, the emissions from smokestacks of major point sources, such as power plants, may enter the atmosphere at night above the nocturnal inversion directly into the nighttime free troposphere.

Nitrogen oxide and VOC emissions continue to react with ozone at night leading to additional reactions involving the nitrate radical. These processes can significantly affect the reactivity and concentration of the pollutant mix that populates the atmosphere during the next diurnal cycle. For these reasons, it is important to better understand the chemistry and transport in the nighttime atmosphere.

Boundary Layer Dynamics: Currently, the temporal and spatial variability of turbulent mixing is the primary challenge to our understanding of planetary boundary layer dynamics. The depth of mixing depends in complex ways on the time of day, the inhomogeneous surface, and the previous state of the atmosphere upwind. We need to better understand the processes that govern the mixing depth on small (2-5 km) and medium (10-50 km) scales. These processes are largely driven by the details of the surface moisture availability and vegetation. We also need to understand the afternoon transition from the vigorously mixed daytime convective boundary layer to the stratified or weakly mixed nighttime residual layer. The transition characteristics may depend on

(aerosol) radiative forcing within the boundary layer as well as on forcing from the surface. Another challenge is to understand the role of clouds in mixing and ventilating the boundary layer. Finally, we need to determine whether the highly intermittent and spatially variable nighttime stable boundary layer has important effects on regional air quality, and if so, how to adequately measure and model this layer.

New Technologies – Investments in the Future

Our ability to understand, and therefore predict, atmospheric processes that control the production and distribution of air pollutants like ozone and PM is measurement-limited. In other words, the advancement of the science is impeded by our inability to properly characterize the state of the atmosphere in terms of chemical composition and physical characteristics. Several examples of technology development efforts that target key parameters whose determination can significantly advance our understanding are described below.

Semi-continuous aerosol speciation: Unlike other air pollutants, such as ozone, PM is not a single compound but rather a complex mixture whose composition and morphology can vary in time and space. Our current knowledge of atmospheric PM stems primarily from 24-hr filter samples. Measurements that resolve the chemical composition of bulk aerosols and individual particles on shorter time scales are required if we are to understand the association between ambient PM and adverse impacts to public health, visibility, and climate forcing. Semi-continuous aerosol speciation will also aid in the proper apportionment of measured PM mass among the myriad of possible emission sources. Thus, the development, enhancement, and evaluation of techniques to speciate ambient aerosols on a semi-continuous basis are a priority.

Chemical Ionization Mass Spectrometry (CIMS): The Aeronomy Laboratory has successfully applied the CIMS technology to the development of a high-sensitivity, continuous nitric acid detector. This technology shows great promise, with the potential for fast-response (one second or less), ultra-sensitive (part-per-trillion) detection of a wide range of compounds of atmospheric interest. Plans call for the development of CIMS-based measurement technologies for the following compounds and classes of compounds:

- Ammonia (NH_3) – Ammonia plays a key role in the formation of fine particles in the atmosphere, the formation of regional haze, the neutralization of acid aerosols, and in acid deposition. No reliable continuous measurement technology currently exists for NH_3. Ammonia measurements can provide insight into gas-to-particle conversion and the impact that these processes may have on air quality, atmospheric visibility, acidic deposition, and human health.
- Isoprene – Naturally produced isoprene is ubiquitous to the temperate regions of the country, where it plays a critical role in ozone formation. Measurements of isoprene and its spatial and temporal distribution will allow a better estimate of the role of natural emissions in ozone formation in rural and urban areas.

- Aromatics – Aromatic compounds, found primarily in automobile emissions, are extremely reactive in the atmosphere, contributing both to ozone formation and the formation of particulate matter. Our ability to understand the role of these compounds is impacted by a lack of reliable measurements.
- Carbonyls – Carbonyl compounds are both the products of atmospheric photochemistry and a source of free radicals in the atmosphere. Thus, they are important indicators of atmospheric reactivity and their quantification can provide insight into atmospheric processes leading to ozone and PM formation.

Aerosol measurement methodologies: Remote optical sensing holds great promise for measuring properties of aerosol particles related to air quality and visibility. Instruments such as lidars and spectral sun photometers can provide continuous information on aerosols aloft, whereas *in situ* measurements by aircraft are restricted to relatively short flight periods, one altitude at a time. The greatest opportunity lies not so much in improving the instruments, which individually are quite mature, but rather in developing new sampling and analysis methodologies that use multiple instruments in a synergistic manner.

Some promising avenues that should be pursued and evaluated include:

- Simultaneous lidar and sun photometer data to characterize column-average size distribution, and major changes of the latter with height.
- Measurement of the profile of water in the aerosols using one or two lidar wavelengths which fortuitously may be only weakly sensitive to variation of other parameters of the size distribution.
- Study of f(RH) throughout the boundary layer
- Continuous remote profiling of $PM_{2.5}$.
- Use of lidar depolarization ratio, which depends on the degree of nonsphericity of the particles, to indicate aerosol formation processes.
- Development of a lidar method to profile ammonia. The lidar could also be used to characterize the strength of point or area sources of ammonia.

Continuous remote profiling of ozone concentration, moisture, and aerosol properties:
Radar wind profilers provide valuable information on the wind and temperature fields in support of air quality research. Technology advancements in optical components now make it realistic to develop and deploy compact, ground-based remote sensors for continuous profiling of ozone, water vapor, and aerosol properties in the lower troposphere. The planned instruments will incorporate solid-state laser technology for compactness and reliability, and will be designed to operate autonomously for extended periods to support both intensive field experiments and long-term monitoring efforts. Analogous with wind profilers, the proposed systems will be inexpensive and easily deployable, so that multiple instruments can conceivably be sited at several locations across a regional area of interest.

Remote sensors for aircraft deployment: The capability to remotely sense boundary layer quantities from aircraft such as the NOAA P-3 and Twin Otter would greatly enhance field studies associated with regional air quality and other Health of the Atmosphere

issues. Lidar remote sensors suitable for aircraft deployment could provide important information on the following quantities:

- Lower tropospheric wind profiles and shear, and the role of inertial oscillations in the transport of urban and power plant plumes.
- Three-dimensional structure of water vapor and aerosols, and the correlation between humidity and aerosol scattering.
- Growth and height of the mixed layer, regional variability of mixed layer height, and the correlation between mixed layer height and surface temperature.
- Three-dimensional variability of ozone across metropolitan areas.
- Turbulent mixing, entrainment, and cloud venting of aerosols and other pollutants.

To provide airborne observational capabilities, technologies already demonstrated at NOAA for surface observations will be modified and extended. Appropriate scanning techniques, including correction of airborne wind observations for aircraft motion, will also need to be investigated. Ultimately, developing instruments for P-3 deployment will further NOAA's missions in areas such as hurricane tracking and forecasting and severe weather research, as well as greatly aiding future Health of the Atmosphere field investigations.

Field Studies – The "Real" World

Intensive regional field studies conducted jointly with colleagues from the Southern Oxidants Study will continue to be a major focus of Health of the Atmosphere research. These studies provide an opportunity to better understand the processes that control the formation and distribution of ozone and fine particles and to develop and test relevant hypotheses. Experience from previous studies has demonstrated the extreme value of three dimensional information provided by remote sensing techniques and instrumented aircraft capable of making a wide range of *in situ* chemical and physical measurements. Two major field campaigns are planned for the 1999- 2000 timeframe.

Southeastern U.S.: NOAA led a collaborative effort that involved several other federal agencies (DOE, EPA, TVA, NSF and NPS), researchers from 10 universities, and industry scientists in a major field campaign conducted in the southeastern U.S. during the summer of 1999. Four instrumented aircraft, including the NOAA WP-3 and a commercial aircraft leased by NOAA as a platform for the ozone aerosol lidar, and a comprehensive array of ground-based measurements were deployed in the region. The study had a regional focus and contrasted and compared the impact of two urban centers, Nashville, Tennessee and Atlanta, Georgia, on local and regional air quality. This study afforded the opportunity to refine our understanding of ozone formation and to begin to address the complex process that determine PM formation and composition. The evaluation of new technologies for PM characterization was a major goal of the field campaign.

The NOAA WP-3 also performed measurements in the Upper Midwest to provide contrasting data from regions with different meteorology and emission profiles.

East Texas: Texas has several challenging air quality problems. Four of the major metropolitan areas have been unable to comply with the National Ambient Air Quality Standard (NAAQS) for ozone, with Houston recording some of the highest ozone concentrations measured anywhere in the country. Texas also has problems meeting air quality standards for particulate matter and regional haze. These problems are compounded by the varied nature of the Texas landscape and climatology.

NOAA will have a leadership role in a study to be conducted in East Texas in the summer of 2000. The study will involve many of the same SOS colleagues that who have worked together on previous studies as well as local scientists from universities, the regulatory community, and industry. The emissions and meteorology that characterize this part of the country are expected to provide a significant departure from the conditions that typified previous studies in the Southeast. Data collected during the study will test the predictive ability of air quality models under a wide range of conditions.

The East Texas study will afford the opportunity to investigate important new research questions:

- *Atmospheric Transport* - Ozone concentrations in Houston are highest when an airmass recirculation occurs due to the presence of a land/sea breeze coupling with the Gulf of Mexico. Under these conditions, pollution from Houston moves toward the Gulf and then returns as the flow changes from a land breeze to a sea breeze. A better understanding of this phenomenon will lead to more effective management strategies.

- *Tracer studies* - Establishing the connection between changes in emissions and the ambient concentrations of the compounds contained in those emissions is a formidable task. The most straightforward approach is through the use of time-series analyses of a long-term record of ambient concentrations of compounds to isolate a signal that can be associated with emission management strategies. However, this approach presents a variety of challenging problems. The time-series record must encompass periods significantly before and after initiation of new air-quality management approaches. The data set must be composed of highly accurate and precise measurements. Even under those circumstances, identification of a shift in the time series of the quantity of interest can be obscured by other transient factors (e.g., meteorological variations or implementation of other emission control programs). Thus, there is a need to develop and evaluate techniques for detecting ambient effects for a specific control program separately from the effects of meteorological variability.

  For these reasons, it is recommended that an alternative approach is recommended to document the effect of various emission control programs on ambient ozone and fine particle precursor concentrations. This alternate approach would use measurements of various tracers in conjunction with measurements of VOC, CO, and NO_x, etc., to (i) characterize the contributions of targeted emissions to the concentrations of ozone- and aerosol precursor compounds; (ii) estimate the ozone- and aerosol-forming

potential of these compounds through the application of various observation-based methods, and (iii) document the change in this contribution that can be attributed to the emission control program. Tracer species that will be useful in this regard include those that can be used to identify light duty vehicle (LDV) emissions (e.g., acetylene for LDV exhaust), as well as those that can serve as a fingerprint of emissions from LDVs using reformulated gasoline (e.g., MTBE). Ideally, these measurements would be made in a variety of locations within and surrounding each severe nonattainment area in order to document effects occurring on regional as well as local and urban scales.

- *Aerosol Formation and Composition* – The presence of extensive petrochemical processing facilities, and the interaction of urban pollution with marine aerosols, is expected to result in an aerosol mix in East Texas unlike anything observed in previous SOS studies. The characterization of these aerosols in terms of their composition and an understanding of the atmospheric processes responsible for their formation and distribution will aid in quantifying public exposure and the development of mitigation strategies.

The NOAA ozone/aerosol lidar was carried aboard a deHavilland Caribou aircraft (top), during the 1999 SOS Nashville field campaign. This aircraft is specially configured for remote sensing studies with a crew of two or three scientists to operate the lidar, evaluate the measurements, and direct the mission (bottom). The airborne lidar provided three dimensional measurements of ozone and fine particle concentrations over the Nashville area (see pages 16-18), enabling researchers to track the evolution and transport of these pollutants throughout the region.

APPENDIX A
HEALTH OF THE ATMOSPHERE PUBLICATIONS

1999

Golden, P.D., D.D. Parrish, W. Kuster, M. Trainer, S. McKeen, J. Holloway, T. Jobson, D. Sueper, and F.C. Fehsenfeld, Airborne measurements of isoprene, CO, and anthropogenic hydrocarbons and their precursors, *Journal of Geophysical Research*, in press, 1999.

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APPENDIX B
LIST OF ACRONYMS AND CHEMICAL SYMBOLS

Acronyms

AHC	Anthropogenic (man made) hydrocarbon
AIRMoN	Atmospheric Integrated Research Monitoring Network
AL	Aeronomy Laboratory (NOAA)
ARL	Air Resources Laboratory (NOAA)
ASL	above sea level
BHC	biogenic hydrocarbon
CASTNet	Clean Air Status and Trends Network
CDT	central daylight time
CENR	Committee on the Environment and Natural Resources
CIMS	chemical ionization mass spectrometry
CIRES	Cooperative Institute for Research in Environmental Sciences
CMDL	Climate Monitoring and Diagnostics Laboratory (NOAA)
DDIM	dry deposition inferential method
DIAL	differential-absorption lidar
DOE	Department of Energy
EPA	Environmental Protection Agency (U.S.)
ETL	Environmental Technology Laboratory (NOAA)
FY	fiscal year
h	mixing height
km	kilometer
LES	large eddy simulation
m	meters
NAAQS	National Ambient Air Quality Standard
NADP	National Acid Deposition Network
NARSTO	a public/private partnership focused on ozone and PM research (formerly known as the North American Research Strategy for Tropospheric Ozone)
nm	nanometer (10^{-6} meter)
NOAA	National Oceanic and Atmospheric Administration
NPS	National Park Service
NSF	National Science Foundation
NTN	National Trends Network
NW	northwest
PBL	planetary boundary layer
PM	particulate matter
$PM_{2.5}$	PM with aerodynamic diameter less than 2.5 microns
ppbv	parts per billion by volume
RH	relative humidity
s	second
SE	southeast
SOS	Southern Oxidants Study

TVA	Tennessee Valley Authority
UV	ultra violet
V_d	deposition velocity

Chemical Symbols

CO	carbon monoxide
HNO_3	nitric acid
HO	hydroxyl radical
MPAN	peroxymethacrylic nitric anhydride
NH_3	ammonia
NMHC	non-methane hydrocarbon
NO_x	nitrogen oxides
O_3	ozone
PAN	peroxy acetic nitric anhydride
PPN	peroxypropionic nitric anhydride
ROO	alkyl-peroxy radical
SO_2	sulfur dioxide
VOC	volatile organic compound

HEALTH OF THE ATMOSPHERE

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