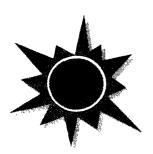
United States Environmental Protection Agency Office of Air and Radiation Washington D C 20460

EPA 452/R-97-002 August 1997

Air

EPA

# Nitrogen Oxides: Impacts on Public Health and the Environment



Stratospheric Ozone Depletion



Global Warming



**Toxic Products** 



Ozone, Particulate Matter, and Nitrogen Dioxide



Acid Deposition



Visibility, Drinking Water and Ecosystem Protection



Eutrophication

# Nitrogen Oxides: Impacts On Public Health and the Environment

Office of Air and Radiation United States Environmental Protection Agency

August 1997

#### Acknowledgments

This report was prepared by a team within the United States Environmental Protection Agency's (EPA) Office of Air and Radiation and was coordinated with EPA's Office of Water. The principal author of this report was Doug Grano, Office of Air Quality Planning and Standards (OAQPS), Air Quality Strategies and Standards Division, Research Triangle Park NC 27711. Authors of specific sections were:

Doris Price and Rona Birnbaum Office of Atmospheric Programs Acid Rain Division, EPA 401 M Street S.W. Washington D.C. 20460 Acid Deposition section;

Richard Batiuk, Chesapeake Bay Program Office, EPA 410 Severn Avenue, Suite 109 Annapolis MD 21403 and Melissa McCullough, OAQPS, EPA Research Triangle Park NC 27711 Eutrophication section; and

Dr. Roy Smith, OAQPS, EPA Research Triangle Park NC 27711 *Toxic Products* section.

Additional contributors to this report were as follows: John Ackermann (EPA, OAQPS); John Bachmann (EPA, OAQPS); Dr. M. Robins Church (EPA, National Health and Environmental Effects Research Lab, Corvallis, OR); Stephen Clark (EPA, Office of Ground Water and Drinking Water); Ted Creekmore (EPA, OAQPS); Robin Dunkins (EPA, OAQPS); Chybryll Edwards (EPA, OAQPS); Barry Gilbert (EPA, OAQPS); Margaret Kerchner (National Oceanic and Atmospheric Administration, Chesapeake Bay Office); Arnold Kuzmack, (EPA, Office of Science and Technology); Chris Lindhjem (EPA, Office of Mobile Sources); Ned Meyer (EPA, OAQPS); Roberta Parry (EPA, Office of Policy Planning and Evaluation); Bruce Polkowsky (EPA, OAQPS); Dr. Doug Ryan (United States Forest Service) and Eric Slaughter (EPA, Oceans and Coastal Protection Division, Office of Wetlands, Oceans, and Watersheds).

This document was reviewed by the following scientific experts: Dr. Ellis Cowling (North Carolina State University), Dr. Rudolph Husar (Washington University at St. Louis, MO), Dr. Harvey Jefferies (University of North Carolina, Chapel Hill), Dr. Hans Paerl (University of North Carolina, Chapel Hill), Dr. Richard Stolarski (National Aeronautics and Space Administration), and Dr. Mark Utell (University of Rochester Medical Center, NY). Their constructive comments provided critical assistance to improving the scientific perspective of the final report.

Finally, public comments were invited on the November 1996 draft of the document; discussion of the document and invitation to comment were provided through the Ozone Transport Assessment Group's Implementation Strategies and Issues Workgroup and at the December 5, 1996 Clean Air Act Advisory Committee meeting. The final document includes consideration by the authors of the comments received from the public. Comments were received from: Colorado State University, Desert Research Institute, Houston Lighting & Power, Pacific Enterprises, Southern Company, Henry L. Stadler, and United States Forest Service.

Copies of this report may be downloaded from the Office of Air and Radiation Policy and Guidance website at http://www.epa.gov/ttn. Questions regarding this report should be directed to Doug Grano at (919) 541-3292.

# Nitrogen Oxides: Impacts on Public Health and the Environment

Table of Contents

			<u>Page</u>
	Exect	utive Summary	1
I.	Introduction/Overview		8
II.	Clean Air Act Programs Involving Decreases in Nitrogen Oxides(NO <sub>x</sub> )Emissions		
	A.	Acid Deposition	15
	в.	Nitrogen Dioxide	31
	C.	Ozone	36
	D.	Particulate Matter	52
	Ε.	Visibility Protection	65
III.	. Additional Public Health and Environmental Impacts from $\mathrm{NO}_{\mathrm{x}}$ Emissions		
	Α.	Drinking Water	75
	в.	Eutrophication	79
	C.	Global Warming	93
	D.	Stratospheric Ozone Depletion	100
	Ε.	Terrestrial Ecosystems	104
	F.	Toxic Products	109
IV.	Interprogram Issues		
	Α.	Local and Regional $NO_x$ Controls	112

В.	Timing of NO <sub>x</sub> Emissions Reductions: Seasonal or Year-Round	120
C.	Interface with Other Control Programs: Three Examples of Secondary Emissions, EPA's Clean Air Power Initiative, and New Standards for Ozone and Particulate Matter	126
Appendice	5	
Α.	Mobile Source Programs	134
в.	Stationary Source Programs	140
C.	Sources and Sinks of Atmospheric Nitrogen	146

D. Acronyms and Abbreviations 155

# Figures and Tables

		<u>Page</u>
Figure I-1	National Anthropogenic NO <sub>x</sub> Emissions by Source Category for 1994 and 2000	10
Figure I-2	National Total NO <sub>x</sub> Emissions by Source Category for 1990	11
Figure I-3	Trends in NO <sub>x</sub> Emissions for the Period 1940 to 1994	13
Figure II-1	Map of Counties Potentially Not Meeting the 8-Hour Ozone Standard	39
Figure II-2	Map of Areas Not Meeting the 1-Hour Ozone Standard	40
Figure II-3	Map of Areas Not Meeting the Particulate Matter $(PM_{10})$ Standard	54
Figure II-4	Yearly Average Absolute and Relative Concentrations for Sulfate and Nitrate	59
Figure III-1	Greenhouse Gas Emissions	94
Figure III-2	Nitrous Oxide Emission Sources	96
Figure C-1	Plants in 1990 with Greater Than 1,000 Tons Per Year of NO <sub>x</sub> Emissions	147
Figure C-2	Density Map of 1994 NO <sub>x</sub> Emissions	148
Figure C-3	Trends in On-Road NO <sub>x</sub> Emissions, Vehicle Miles Traveled, Fuel Use, and Gasoline Price	150

Table II-1	Utility Boiler Types and Emission Limits	17
Table II-2	Constituents of Atmospheric Fine Particles Less Than 2.5 Microns and Their Major Sources	61
Table II-3	Constituents of Atmospheric Particles Greater Than 2.5 Microns and Their Major Sources	62
Table II-4	Annual Averages (March 1988-February 1991) of Reconstructed Light Extinction (Mm <sup>-1</sup> ) for 19 Regions of the IMPROVE Network	72
Table III-1	Estimated Reductions in Nitrogen Loadings to Chesapeake Bay and Water Quality Response Under Several Control Scenarios	89
Table III-2	Estimated global sources of Nitrous Oxide	97
Table IV-1	Seasonal Emissions of NO <sub>x</sub> , 1985 through 1994	124
Table C-1	1994 National NO <sub>x</sub> Emissions by Source Category	149
Table C-2	Total National Emissions of NO <sub>x</sub> , 1940 through 1994	151
Table C-3	Estimated Global Emissions of NO <sub>x</sub> Typical of the Last Decade	152
Table C-4	Estimated Global Emissions of NH <sub>3</sub>	153

# Nitrogen Oxides: Impacts On Public Health and the Environment

# Executive Summary

#### **Overview:**

Over the past two decades, great progress has been made at the local, state and national levels in controlling emissions from many sources of air pollution. However, pollutant levels remain unacceptably high in many areas across the country. The Clean Air Act (CAA) specifies deadlines for attainment of the ozone ( $O_3$ ) standards, yet continued industrial growth and expansion of motor vehicle usage threaten to reverse past achievements. An abundance of  $O_3$  near the earth's surface results in damaging effects on human health, agricultural crops, ornamental plants, forests, and materials.

For many years, control of volatile organic compounds (VOCs) was the main strategy employed in efforts to decrease ground-level  $O_3$ . More recently, it has become clearer that decreases in emissions of nitrogen oxides (NO<sub>x</sub>) may be needed in many areas, especially in areas where  $O_3$  concentrations are high over a large region (as in the Midwest, Northeast, and Southeast). The 1991 National Academy of Sciences report entitled <u>Rethinking the Ozone</u> <u>Problem in Urban and Regional Air Pollution</u> recommends that "To substantially reduce  $O_3$ concentrations in many urban, suburban, and rural areas of the United States, the control of  $NO_x$ emissions will probably be necessary in addition to, or instead of, the control of VOCs."

In addition to attainment of the public health standards for  $O_3$ , decreases in emissions of  $NO_x$  are helpful to several other efforts to improve the environment. On a national scale, decreases in  $NO_x$  emissions will also decrease acid deposition, nitrates in drinking water, excessive nitrogen loadings to aquatic and terrestrial ecosystems, and ambient concentrations of nitrogen dioxide, particulate matter and toxics. On a global scale, decreases in  $NO_x$  emissions will, to some degree, reduce greenhouse gases and stratospheric  $O_3$  depletion. Thus, management of air emissions is essential to both air quality and watershed protection on national and global scales.

In view of the need for  $NO_x$  emissions decreases in the  $O_3$  program and the multiple environmental benefits that would follow, EPA's Office of Air and Radiation, in coordination with EPA's Office of Water, has begun implementing an integrated approach to achieve substantial decreases in the emissions of  $NO_x$  from mobile and stationary sources. In particular, EPA's Offices of Air Quality Planning and Standards, Atmospheric Programs, and Mobile Sources are implementing this strategy by taking a balanced approach to decreasing  $NO_x$  emissions among several categories of mobile and stationary sources, considering costs, effectiveness, alternatives, and opportunities for market incentives. This integrated approach involves increased interaction among the air and water programs that are affected by various forms of atmospheric nitrogen. This interaction is needed so that implementation of the  $NO_x$  emissions decreases occurs in a manner that best achieves the multiple public health and environmental goals. Thus, policy decisions regarding the control of  $NO_x$  emissions are being made in the context of the many environmental effects associated with  $NO_x$  emissions.

### Multiple Public Health and Environmental Benefits Flow from NO<sub>x</sub> Emissions Decreases

The impact of NO<sub>x</sub> emissions on O<sub>3</sub> concentrations is complex. Although NO<sub>x</sub> emissions are necessary for the formation of O<sub>3</sub> in the lower atmosphere, a local decrease in NO<sub>x</sub> emissions can, in some cases, increase local O<sub>3</sub> concentrations. This effect of NO<sub>x</sub> emissions decreases must be carefully weighed against the multiple benefits than can be associated with decreasing NO<sub>x</sub> emissions, including lowering regional O<sub>3</sub> concentrations. It should be noted that, with EPA's July 18, 1997 promulgation of the new O<sub>3</sub> standards, greater emphasis might be needed on regional-scale NO<sub>x</sub> emissions decreases to reach attainment because the new standards result in more areas and larger areas with monitoring data indicating nonattainment. Specifically, NO<sub>x</sub> emissions also contribute to adverse impacts to public health and the environment in the following areas:

Acid Deposition: Sulfur dioxide and  $NO_x$  are the two key air pollutants that cause acid deposition (wet and dry particles and gases) and result in the adverse effects on aquatic and terrestrial ecosystems, materials, visibility, and public health. Nitric acid deposition plays a dominant role in the acid pulses associated with the fish kills observed during the springtime melt of the snowpack in sensitive watersheds and recently has also been identified as a major contributor to chronic acidification of certain sensitive surface waters.

**Drinking Water Nitrate**: High levels of nitrate in drinking water is a health hazard, especially for infants. Atmospheric nitrogen deposition in sensitive watersheds can increase stream water nitrate concentrations; the added nitrate can remain in the water and be transported long distances downstream.

**Eutrophication**:  $NO_x$  emissions contribute directly to the widespread accelerated eutrophication of United States coastal waters and estuaries. Atmospheric nitrogen deposition onto surface waters and deposition to watershed and subsequent transport into the tidal waters has been documented to contribute from 12 to 44 percent of the total nitrogen loadings to United States coastal waterbodies. Nitrogen is the nutrient limiting growth of algae in most coastal waters and estuaries. Thus, addition of nitrogen results in accelerated algae and aquatic plant growth causing adverse ecological effects and economic impacts that range from nuisance algal blooms to oxygen depletion and fish kills.

**Global Warming**: Nitrous oxide  $(N_2O)$  is a greenhouse gas. Anthropogenic  $N_2O$ 

emissions in the United States contribute about 2 percent of the greenhouse effect, relative to total United States. anthropogenic emissions of greenhouse gases. In addition, emissions of  $NO_x$  lead to the formation of tropospheric  $O_3$ , which is another greenhouse gas.

**Nitrogen Dioxide** (NO<sub>2</sub>): Exposure to NO<sub>2</sub> is associated with a variety of acute and chronic health effects. The health effects of most concern at ambient or near-ambient concentrations of NO<sub>2</sub> include mild changes in airway responsiveness and pulmonary function in individuals with pre-existing respiratory illnesses and increases in respiratory illnesses in children. Currently, all areas of the United States monitoring NO<sub>2</sub> are below EPA's threshold for health effects.

**Nitrogen Saturation of Terrestrial Ecosystems**: Nitrogen accumulates in watersheds with high atmospheric nitrogen deposition. Because most North American terrestrial ecosystems are nitrogen limited, nitrogen deposition often has a fertilizing effect, accelerating plant growth. Although this effect is often considered beneficial, nitrogen deposition is causing important adverse changes in some terrestrial ecosystems, including shifts in plant species composition and decreases in species diversity or undesirable nitrate leaching to surface and ground water and decreased plant growth.

**Particulate Matter (PM)**:  $NO_x$  compounds react with other compounds in the atmosphere to form nitrate particles and acid aerosols. Because of their small size nitrate particles have a relatively long atmospheric lifetime; these small particles can also penetrate deeply into the lungs. PM has a wide range of adverse health effects.

**Stratospheric O<sub>3</sub> Depletion**: A layer of O<sub>3</sub> located in the upper atmosphere (stratosphere) protects people, plants, and animals on the surface of the earth (troposphere) from excessive ultraviolet radiation. N<sub>2</sub>O, which is very stable in the troposphere, slowly migrates to the stratosphere. In the stratosphere, solar radiation breaks it into nitric oxide (NO) and nitrogen (N). The NO reacts with O<sub>3</sub> to form NO<sub>2</sub> and molecular oxygen. Thus, additional N<sub>2</sub>O emissions would result in some decrease in stratospheric O<sub>3</sub>.

**Toxic Products**: Airborne particles derived from  $NO_x$  emissions react in the atmosphere to form various nitrogen containing compounds, some of which may be mutagenic. Examples of transformation products thought to contribute to increased mutagenicity include the nitrate radical, peroxyacetyl nitrates, nitroarenes, and nitrosamines.

**Visibility and Regional Haze**:  $NO_x$  emissions lead to the formation of compounds that can interfere with the transmission of light, limiting visual range and color discrimination. Most visibility and regional haze problems can be traced to airborne particles in the atmosphere that include carbon compounds, nitrate and sulfate aerosols, and soil dust. The major cause of visibility impairment in the eastern United States is sulfates, while in the West the other particle types play a greater role.

#### **O**<sub>3</sub> Formation and Accumulation

Although  $O_3$  formation and accumulation in the atmosphere involves complex nonlinear processes, a very simplified description of the process is offered here. In short, NO is formed during high temperature combustion involving air (air being largely  $N_2$  and  $O_2$ ). The NO is converted to  $NO_2$  by reacting with either inorganic or organic radicals formed from oxidized VOCs or by reacting with  $O_3$ . The  $NO_2$  then photolyzes, leading to the formation of  $O_3$  and NO. A reaction path that converts NO to  $NO_2$  without consuming a molecule of  $O_3$  allows  $O_3$  to accumulate; such a path is provided by inorganic and organic radicals that arise from VOC reactions.

The formation and accumulation of  $O_3$  is further complicated by the transport of  $O_3$  itself and  $O_3$  precursors (including  $NO_x$ ). This transport factor results in interactions between distant sources in urban or rural areas and local ambient  $O_3$  concentrations. The transport of  $O_3$  and precursor pollutants over hundreds of kilometers (or hundreds of miles) can be a significant factor in the accumulation of  $O_3$  in certain areas. Another important complicating factor is the influence of meteorological factors on  $O_3$  formation, including temperature, wind direction, and wind speed.

In the 1990 amendments to the CAA, Congress recognized the importance of  $NO_x$  emissions reductions and, especially in the Northeast, the need for regional scale control programs to achieve the  $O_3$  standard. In section 184 of the CAA, Congress established the Northeast Ozone Transport Commission to address interstate transport of  $O_3$  pollution among 12 northeastern States and the District of Columbia. Further, Congress required large stationary sources located in the Northeast Ozone Transport region and in moderate, serious, severe and extreme  $O_3$  nonattainment areas throughout the country to decrease  $NO_x$  emissions.

The extent of local controls that will be needed to attain and maintain the  $O_3$  national ambient air quality standards (NAAQS) in and near seriously polluted cities is sensitive both to the amount of  $O_3$  and  $O_3$  precursors transported into the local area and to the specific photochemistry of the area. In some cases, preliminary local modeling performed by the states for the 1-hour  $O_3$ standard indicates that it may not be feasible to find sufficient local control measures for individual nonattainment areas unless transport into the areas is significantly lowered. The EPA has also conducted preliminary analyses for the new 8-hour  $O_3$  standard which indicate that regional  $NO_x$ emissions decreases would be effective in helping many areas attain that standard. These modeling studies suggest that decreasing  $NO_x$  emissions on a regional basis is effective in decreasing  $O_3$  over large geographic areas.

## NO<sub>x</sub> Emissions Sources and Trends

Emissions of  $NO_x$  result from fuel combustion at high temperature, which occurs principally in fossil fuel-fired electric utility and industrial boilers and in motor vehicle internal combustion engines. Electric utility and motor vehicle emissions each represent about one-third of the total 1994  $NO_x$  emissions. About 85 percent of the total  $NO_x$  emissions from electric utilities are attributed to utilities burning coal. From 1940 through 1970, annual  $NO_x$  emissions increased by a factor of three (from 7 million to 21 million tons). Since 1980, annual national  $NO_x$  emissions leveled off at about 23 million tons. Data show that national  $NO_x$  emissions slightly increased from 1990-1993. In the mid-1990s,  $NO_x$  emissions are expected to decrease somewhat as stationary source  $NO_x$  controls and light-duty and heavy-duty tailning standards are implemented and enhanced vehicle inspection

and light-duty and heavy-duty tailpipe standards are implemented and enhanced vehicle inspection and maintenance (I/M) programs begin in some  $O_3$  nonattainment areas. Electric utility  $NO_x$ emissions are expected to decline after 1999 as the phase II acid deposition standards become effective. Despite increases in vehicle miles traveled, total on-road vehicle emissions will likely continue to decline through 2005 as per vehicle  $NO_x$  emissions decrease due to tighter tailpipe standards, phase II reformulated gasoline is implemented, and I/M requirements are met. Soon after the year 2002, overall  $NO_x$  emissions are projected to begin to increase and continue to increase in the foreseeable future due to increased economic activity.

#### General Conclusions and Implications for Future NO<sub>x</sub> Management Strategies

It has become clearer that controls of  $NO_x$  emissions may be needed in many areas, especially in areas of the United States where  $O_3$  concentrations are high over a large region (as in the Midwest, Northeast, and Southeast). In addition to helping attain the NAAQS for  $O_3$ , decreases in  $NO_x$  emissions will also likely help improve the environment by decreasing the adverse impacts of acid deposition, drinking water nitrate exposure, eutrophication of waterbodies, global warming,  $NO_2$  exposure, nitrogen saturation of terrestrial ecosystems, PM formation, stratospheric  $O_3$  depletion, toxics exposure, and visibility impairment.

Although total  $NO_x$  emissions will decline from current levels by the year 2000 because of mandatory CAA programs,  $NO_x$  emissions will, soon after the year 2002, begin to gradually increase. Both mobile, including non-road, and stationary sources are significant contributors to the  $NO_x$  problem on a nationwide basis. Thus, new initiatives will be necessary to achieve reductions in  $NO_x$  emissions that may be needed over much of the nation, especially to help attain the  $O_3$  standards.

The EPA has begun implementing an integrated approach to achieve reductions in emissions of  $NO_x$ . This integrated approach involves increased interaction among the air and water programs that are affected by various forms of atmospheric nitrogen and addresses several categories of mobile and stationary sources. Policy decisions regarding the control of  $NO_x$  emissions are being made in the context of the many environmental effects associated with  $NO_x$  emissions. The EPA continues to work under its own authority and in coordination with a wide range of stakeholders to develop and implement new mobile and stationary source control programs at the federal, state, and local levels to decrease emissions of  $NO_x$ . The following are the key aspects of this strategy:

## Mobile Sources

Since the 1970's EPA has required motor vehicle manufacturers to decrease significantly emissions of  $NO_x$  from light duty on-road vehicles. The most recent light

duty vehicle requirements were phased-in over the 1994-96 model years. The EPA continues to work with state officials, auto manufacturers, oil industry and others to develop even cleaner cars, known as the National Low Emission Vehicles program. Reduction in NO<sub>x</sub> emission levels from heavy-duty vehicles is expected from lower tailpipe standards for engines produced after 1991 and further reductions are expected with the 1998 and 2004 model year engines. In 1995 cities with the worst smog problems in the nation began using cleaner reformulated gasoline; a second phase of that program will reduce emissions of NO<sub>x</sub> beginning in the year 2000. In addition, EPA is working on several non-road programs to decrease NO<sub>x</sub> emissions from large marine, aircraft, locomotive, and general purpose engines like those used in agriculture, construction, and general industrial equipment.

#### Stationary Sources

To help control acid deposition, EPA established a two phased program to reduce emissions of  $NO_x$  from coal-fired electric utility generation units. This program is expected to decrease  $NO_x$  emissions by about 2 million tons annually by the year 2000. States are also requiring controls on large sources of  $NO_x$  that are located in areas of the country that fail to meet the NAAQS for ground-level  $O_3$ . To help decrease ground-level  $O_3$ , twelve northeastern states and the District of Columbia developed a memorandum of understanding to reduce emissions of  $NO_x$  from large boilers by 55-75 percent from 1990 levels. As a means of achieving these reductions with the least cost, EPA is working with these states to develop an emissions trading program.

## Ozone Transport Assessment Group (OTAG)

Over a 2 year period EPA worked with the OTAG, which was chartered by the Environmental Council of States for the purpose of evaluating O<sub>3</sub> transport and recommending strategies for mitigating interstate pollution. The OTAG was a consultative process among 37 eastern states which included examination of the extent that NO<sub>x</sub> emissions from hundreds of kilometers away are contributing to smog problems in downwind cities in the eastern half of the country, such as Atlanta, Boston, and Chicago. The OTAG completed its work in June 1997 and on July 8, 1997 forwarded its recommendations to EPA for achieving additional cost-effective emissions reduction programs to decrease ground-level  $O_3$  throughout the eastern United States. In its recommendations OTAG stated that it recognizes that NO<sub>x</sub> controls for O<sub>3</sub> reduction purposes have collateral public health and environmental benefits, including reductions in acid deposition, eutrophication, nitrification, fine particle pollution, and regional haze. Based on these recommendations and additional information, EPA will complete a rulemaking action requiring States in the OTAG region that are significantly contributing to O<sub>3</sub> nonattainment in downwind States to revise their State implementation plans to include new rules to reduce their emissions of NO<sub>x</sub>.

#### **Emerging Technologies**

Since passage of the 1970 CAA amendments, air pollution control and prevention

technologies have continuously improved. Technologies such as selective catalytic reduction and gas reburn systems are in place and successfully performing today that were only on the drawing board ten years ago. As the demand for more innovative and cost-effective or cost-saving technologies increases-due to the above new initiatives, for example--new technologies such as ultra low-NO<sub>x</sub> gas-fired burners and vacuum insulated catalytic converters will move from the research and development or pilot program phase to commercial availability. Thus, it is likely that many new technologies will be available in the next ten to fifteen years to employ in air pollution control and prevention strategies.

# *Nitrogen Oxides Impacts On Public Health and the Environment*

## I. Introduction/Overview

#### Purpose

The purpose of this document is to describe the multiple impacts on human health and welfare that result from emissions of nitrogen oxides (NO<sub>x</sub>). Emissions of NO<sub>x</sub> result in an unusually broad range of detrimental effects to human health and the environment. In addition, this document states EPA's intent to consider the multiple environmental impacts of NO<sub>x</sub> emissions when making policy decisions regarding regulation of NO<sub>x</sub> emissions.

## Atmospheric nitrogen (N) compounds

Atmospheric N compounds include many forms of N, both inorganic and organic, in gaseous and particulate states. One form of N compound--diatomic N gas  $(N_2)$ --makes up 78 percent of the atmosphere; however, it is inert and, thus, does not readily react with other compounds in the atmosphere. As described below, many important N compounds can be classified as oxidized N or reduced N. Other forms of N compounds are highly reactive and also play a role in the formation and accumulation of various gases and particles in the atmosphere which lead to harmful effects on human health and welfare.

Seven oxides of nitrogen are known to occur in the atmosphere: NO, NO<sub>2</sub>, NO<sub>3</sub>, N<sub>2</sub>O, N<sub>2</sub>O<sub>3</sub>, N<sub>2</sub>O<sub>4</sub>, and N<sub>2</sub>O<sub>5</sub>. "NO<sub>x</sub>" is a symbol for the sum of nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>); these compounds are generally transformed and cycled within the atmosphere through nitrate radical (NO<sub>3</sub>), organic nitrates, and dinitrogen pentoxide (N<sub>2</sub>O<sub>5</sub>), eventually forming nitric acid (NRC, 1991). N<sub>2</sub>O is not formed as part of this atmospheric chemistry of NO<sub>x</sub>. Although not reactive in the lower atmosphere, N<sub>2</sub>O is a significant greenhouse gas which is reactive once it diffuses into the stratosphere. The various forms of oxides of nitrogen--NO<sub>x</sub>, N<sub>2</sub>O, nitrates, etc.-- are discussed separately in this document with respect to specific human health and environmental impacts.

Reduced N compounds--ammonia (NH<sub>3</sub>) and ammonium (NH<sub>4</sub><sup>+</sup>)--are also important to many of the public health and environmental impacts associated with atmospheric N compounds. Additional information on emissions of NH<sub>3</sub> are contained in Appendix C of this document. The emphasis of this report, however, is on oxides of nitrogen--their sources, impacts, and an integrated strategy to decrease their emissions.

#### Anthropogenic NO<sub>x</sub> Emissions Sources

Emissions of  $NO_x$  are produced primarily by combustion processes during which oxygen reacts with nitrogen at temperatures above about 2200 degrees Celsius. Both the molecular N  $(N_2)$  in the atmosphere and the chemically bound N in materials being burned (fuel N) can react with oxygen to form  $NO_x$ . Such combustion occurs principally in fossil fuel-fired electric utility and industrial boilers and in motor vehicle internal combustion engines. As shown in the following chart of anthropogenic emissions (EPA, 1995), electric utility and on-road vehicle emissions each represent about one-third of the total 1994  $NO_x$  emissions (figure I-1). In the year 2000, the percentage of utility emissions is projected to decline as the CAA phase II acid deposition controls are implemented. About 85 percent of the NO<sub>x</sub> emissions category includes marine, aircraft, locomotive and construction equipment. Appendix C contains additional information on anthropogenic  $NO_x$  emissions.

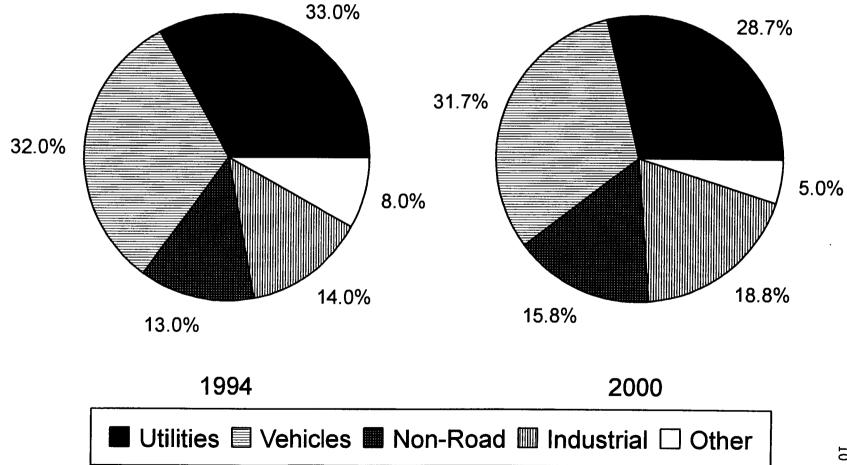
#### Biogenic NO<sub>x</sub> Emissions Sources

Natural sources of NO<sub>x</sub> include lightning, soils, wildfires, stratospheric intrusion, and the oceans. Of these, lightning and soils are the major contributors. Lightning produces high enough temperatures to allow N<sub>2</sub> and O<sub>2</sub> in the atmosphere to be converted to NO. NO is the principal NO<sub>x</sub> species emitted from soils, with emission rates depending mainly on fertilization amounts and soil temperature; highest emissions occur in the summer. The United States 1990 annual biogenic emissions of NO<sub>x</sub> are estimated to be 1.69 million tons (EPA, 1995); using the Biogenic Emissions Inventory System--Version 2. As shown in figure I-2, biogenic emissions are about 7 percent of the total NO<sub>x</sub> emissions in 1990 (EPA, 1995).

In areas with extensive agricultural production, such as the Southeast, biogenic emissions from soil treated with nitrate-rich fertilizer can represent a measurable portion of total  $NO_x$  emissions. Much of the spatial difference in biogenic  $NO_x$  emissions across the United States can be attributed to variations in land use. Relatively high densities of  $NO_x$  in the midwestern United States are associated with areas of fertilized crop land.

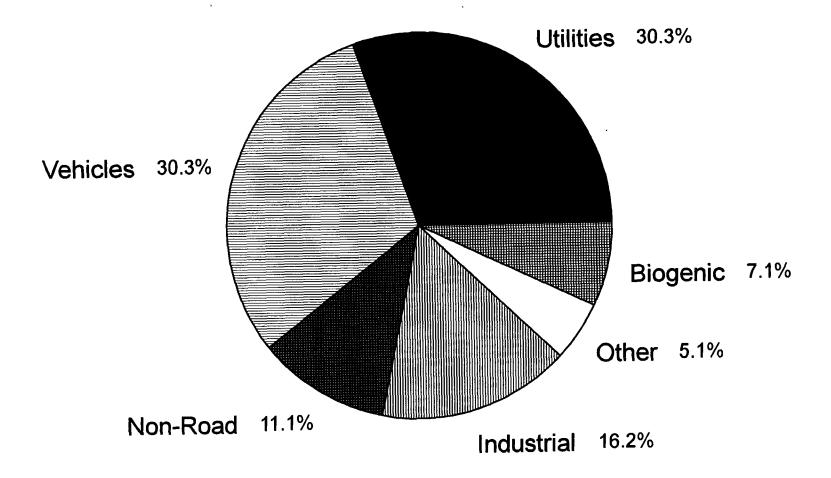
Soil emissions of NO result from two major microbial processes: nitrification and denitrifications. Nitrification is the process by which microbes in the soil oxidize the ammonium ion to produce nitrites and nitrates. During the intermediate stages of this process, NO is formed and subsequently diffuses through the soil into the atmosphere. By contrast, denitrification is an anaerobic process where nitrate is converted to  $N_2$  and  $N_2O$ ; but once again, NO is formed in an intermediate stage and diffuses to the atmosphere. Once in the atmosphere, NO begins to participate in atmospheric chemical reactions. Within a time of tens to hundreds of seconds, a substantial portion of NO has reacted with atmospheric  $O_3$  to produce NO<sub>2</sub> (Aneja, 1994).

# Figure I-1. National Anthropogenic NO<sub>x</sub> Emissions by Source Category for 1994 and 2000



10

# Figure I-2. National Total NOx Emissions by Source Category for 1990

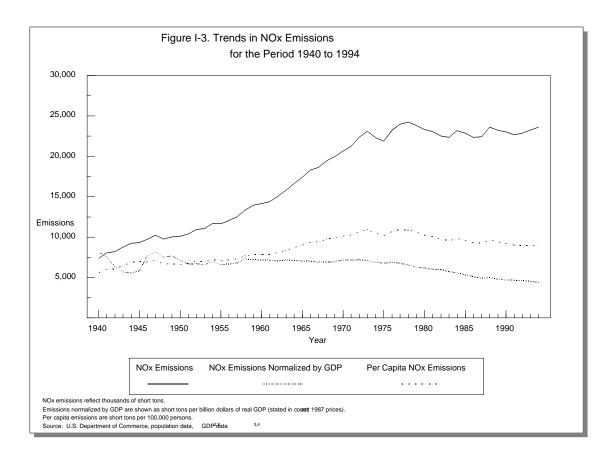


#### Trends in Anthropogenic NO<sub>x</sub> Emissions

From 1940 through 1970,  $NO_x$  emissions increased by a factor of three (from 7 million to 21 million tons). Since 1980, annual national  $NO_x$  emissions have leveled off at about 23 million tons.  $NO_x$  emissions slightly increased from 1990-1993.  $NO_x$  emissions from electric utilities and on-road vehicles currently contribute about one third each to the national total (approximately 8 million tons each).

In the mid-1990s,  $NO_x$  emissions are expected to decrease somewhat as stationary source  $NO_x$  controls and light-duty and heavy-duty tailpipe standards are implemented and enhanced vehicle inspection and maintenance (I/M) programs begin in some  $O_3$  nonattainment areas. Electric utility emissions are expected to decline after 1999 as the phase II acid deposition standards become effective. Total  $NO_x$  emissions will decline about 6 percent from current levels by the year 2000. Despite increases in vehicle miles traveled, total on-road vehicle emissions will likely continue to decline through 2005 as per vehicle emissions decrease due to tighter tailpipe standards, phase II reformulated gasoline is implemented, and I/M requirements are met. Shortly after the year 2002, overall  $NO_x$  emissions are projected to begin to increase and continue to increase in the foreseeable future due to increased economic activity, unless new  $NO_x$  emissions reduction initiatives are implemented (EPA, 1995).

In general, the per capita  $NO_x$  emissions show a much smaller increase during the 1940 to 1978 period than did the total  $NO_x$  emissions trend. Per capita  $NO_x$  emissions have declined since 1978.  $NO_x$  emissions normalized by real Gross Domestic Product (GDP) declined and then increased during the 1940s but declined thereafter, an indication that fewer  $NO_x$  emissions are released per dollar of real GDP. These points are illustrated in figure I-3 below (EPA, 1995).



## Organization of this Document

This document is organized in 5 major sections: Introduction/Overview, Clean Air Act Programs Involving Decreases in NO<sub>x</sub> Emissions, Additional Public Health and Environmental Impacts from NO<sub>x</sub> Emissions, Interprogram Issues, and Appendices. The introduction/overview section outlines the purpose of the document and provides information on atmospheric N compounds, sources of NO<sub>x</sub> emissions, and trends in emissions of NO<sub>x</sub>. The Programs section covers the impact of NO<sub>x</sub> emissions in each of the following subjects: acid deposition, NO<sub>2</sub>, O<sub>3</sub>, PM, and visibility protection. Drinking water, eutrophication, global warming, stratospheric O<sub>3</sub> depletion, terrestrial ecosystems, and toxics products are covered under the additional public health and environmental impacts section. A subsequent section covers specific issues stemming from interaction among the various programs, including local and regional NO<sub>x</sub> concerns, seasonal controls, interface with the VOCs control program, EPA's Clean Air Power Initiative, and crosscutting issues related to the new standards for O<sub>3</sub> and PM. Finally a set of appendices provides some detail on the EPA activities within the various programs that impact NO<sub>x</sub> emissions, information on sources and sinks of NO<sub>x</sub> emissions, and a listing of acronyms and abbreviations.

# References

Aneja, Viney P., "Workshop on the Intercomparison of Methodologies for Soil NO<sub>x</sub> Emissions: Summary of Discussion and Research Recommendations," *Journal of Air and Waste Management Association*, vol. 44, August 1994.

Chameides, W.L. and E.B. Cowling, *The State of the Southern Oxidant Study (SOS): Policy-Relevant Findings in Ozone Pollution Research, 1988-1994.* North Carolina State University, April 1995.

National Research Council, Committee on Tropospheric Ozone Formation and Measurement, *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, National Academy Press, 1991.

Schlesinger, W. H. and A. E. Hartley, "A Global Budget for Atmospheric NH<sub>3</sub>," *Biochemistry* 15: 191-211, 1992.

U.S. Environmental Protection Agency, *Control Techniques for Nitrogen Oxides Emissions from Stationary Sources--Revised Second Edition*, EPA-450/3-83-002, January 1983.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *National Air Pollutant Emission Trends*, 1900-1994, October 1995.

- II. Clean Air Act Programs Involving Decreases in Nitrogen Oxides  $({\rm NO}_{\rm x})$  Emissions
  - A. Acid Deposition
  - 1. Goals of the Program

The primary goal of the Acid Deposition  $NO_x$  Emission Reduction Program is to decrease the multiple adverse environmental and human health effects of  $NO_x$ , a principal acid deposition precursor that contributes to air and water pollution, by substantially decreasing annual emissions from coal-fired power plants. Electric utilities are a major contributor to  $NO_x$  emissions nationwide: in 1980, they accounted for 30 percent of total  $NO_x$  emissions and, from 1980 to 1990, their contribution rose to 32 percent of total  $NO_x$  emissions. Approximately 85 percent of electric utility  $NO_x$  emissions comes from coal-fired plants.

"Acid deposition" occurs when airborne acidic or acidifying compounds, principally sulfates  $(SO_4^{2-})$  and nitrates  $(NO_3^{-})$ , which can be transported over long distances, return to the earth through rain or snow ("wet deposition"), through fog or cloud water ("cloud deposition"), or through transfer of gases or particles ("dry deposition"). While the severity of the damage depends on the sensitivity of the receptor, acid deposition, according to section 401(a)(1) of the CAA, "represents a threat to natural resources, ecosystems, visibility, materials, and public health."

Since  $NO_x$  emissions from the burning of fossil fuels at electric utility power plants contribute to the formation of ground-level  $O_3$  and nitrate PM in the air, ambient levels of  $NO_2$ and peroxyacetal nitrate (PAN) gases, and atmospheric N deposition, the Acid Deposition  $NO_x$ Emission Reduction Program will also mitigate the negative health and welfare effects described in the other sections of this document. Benefits associated with  $NO_x$  emissions decreases under the Acid Deposition Program include lowering excessive N loadings to N sensitive estuarine or coastal water systems ranging from the Gulf of Maine to North Carolina's Albemarle Pamlico Sound to Florida's Tampa and Sarasota Bays, decreasing  $O_3$  transported into and within  $O_3$ nonattainment areas, decreasing inhalable fine particles, and improving visibility, as well as reducing acid deposition damage to lakes and streams, forests and vegetation, and sensitive materials and structures.

2. Status of the Program

Title IV (Acid Deposition Control) of the CAA specifies a two-stage program for decreasing  $NO_x$  emissions from existing coal-fired electric utility power plants. Analogous to the national allowance program for decreasing sulfur dioxide (SO<sub>2</sub>) emissions, this program is to be

implemented in two phases. Phase I affected units  $(277 \text{ boilers})^1$  are required to meet the applicable annual emission rates beginning with calendar year 1996; Phase II affected units (775 boilers) are required to meet the applicable annual emission rates beginning with calendar year 2000. Implementation of the first stage of the program, promulgated April 13, 1995 (60 FR 18751), will decrease annual NO<sub>x</sub> emissions in the United States by over 400,000 tons per year between 1996 and 1999 (Phase I) and by approximately 1.17 million tons per year beginning in 2000 (Phase II). These reductions are achieved by applying low NO<sub>x</sub> burner (LNB) technology to dry bottom wall-fired boilers and tangentially fired boilers (Group 1).

The second stage of the program, promulgated December 19, 1996 (61 FR 67112), provides for additional annual NO<sub>x</sub> emissions reductions in the United States of approximately 890,000 tons per year beginning in the year 2000 (Phase II). Taken together, the two stages provide for an overall decrease in annual NO<sub>x</sub> emissions in the United States of approximately 2.06 million tons per year beginning in the year 2000. In the second stage of the title IV Program EPA has: (1) determined that more effective low NO<sub>x</sub> burner (LNB) technology is available to establish more stringent standards for Phase II, Group 1 boilers than those established for Phase I; and (2) established limitations for other boilers known as Group 2 (wet bottom boilers, cyclones, cell burner boilers, and vertically fired boilers), based on NO<sub>x</sub> control technologies that are comparable in cost to LNBs.

<sup>&</sup>lt;sup>1</sup> 170 Phase I units known as Table 1 units and 107 Phase II units that have become substitution units. (The 170 Table 1 units are coal-fired units with Group 1 boilers listed in Table 1 of 40 CFR 73.10 (a) of the Acid Rain Program Regulations.)

The following table presents the boiler types affected by this rule, their population, and the  $NO_x$  emission limitations:

Boiler Types	Number of Boilers	Phase II Emission Limits	
Phase II, Group 1 Boilers		(Revised)	
dry bottom wall-fired	308	dry bottom wall-fired: 0.46 lb/mmBtu	
tangential	299	tangential: 0.40 lb/mmBtu	
Group 2 Boilers		(New)	
cell burners	36	cell burners: 0.68 lb/mmBtu	
cyclones > 155 MW	55	cyclones: 0.86 lb/mmBtu	
wet bottoms > 65 MW	26	wet bottoms: 0.84 lb/mmBtu	
vertically fired	28	verticals: 0.80 lb/mmBtu	

Table II-1. Utility Boiler Types and Emission Limits

Utilities can choose to comply with the rule in one of three ways: (1) meet the standard annual emission limitations, (2) average the emissions rates of two or more boilers, which allows utilities to over-control at units where it is technically easier and less expensive to control emissions, or (3) if a utility cannot meet the standard emission limit, it can apply for a less stringent alternative emission limit if it uses the appropriate NO<sub>x</sub> emissions control technology on which the applicable emissions limit is based. Although emission limitations for the Acid Deposition NO<sub>x</sub> Emission Reduction Program are based on "the degree of reduction achievable through the retrofit application of the best system of continuous emission reduction" (section 407(b)(1) of the CAA), the annual averaging period affords sources the flexibility of selecting either continuous or seasonal controls.

In addition, the current rule allows utilities to "early elect" Phase II units with Group 1 boilers into the Phase I program, provided the units demonstrate compliance with the applicable annual emission rate on or before January 1, 1997. As an incentive for early reductions, the rule affects early election units from revisions to the emission limits promulgated in 61 FR 67112 through 2007. EPA has received early election applications for over 250 Phase II units (corresponding to about 43 percent of the Phase II affected Group 1 boiler population). The early election and emissions averaging provisions of the Acid Deposition NO<sub>x</sub> Emission Reduction Program offer flexibility, promote technology development and competition, and provide opportunities to reduce the cost of control.

# 3. Science of $\ensuremath{\text{NO}_{x}}$ and Acid Deposition

The burning of fossil fuels is a major contributor to the formation of  $NO_x$  and thus to atmospheric N deposition. "Atmospheric N deposition" is the process by which N in airborne or atmospheric N compounds is transferred to water, soil, vegetation, and other materials (e.g. buildings, statues, automobiles, etc.) on the earth. While some amount of N deposition can be beneficial for growth of crops and forests, deposition in excess of plant and microbial demand can disturb the soil and water N cycle and can result in acidification of lakes, streams, and soils as well as eutrophication of estuarine and coastal waters bodies (Paerl, 1993) and, more rarely, freshwater ecosystems (Church, 1997:17; Vitousek et al, 1997:10). Eutrophication of estuarine and coastal waters is addressed in section III.B of this document.

As mentioned previously, "acid deposition" involves acidic and acidifying sulfur and N compounds, which can be transported over short and long distances, thus affecting natural resources and materials up to hundreds of kilometers from the sources of precursor emissions  $(SO_2 \text{ and } NO_x)$ . As with NO<sub>x</sub> emissions and O<sub>3</sub> formation, the relationship between precursor emissions and acidity in the atmosphere is complex (NAPAP, 1993:23). Some of these acidic and acidifying compounds are not emitted directly during the burning of fossil fuels; they are formed by chemical conversions in the atmosphere of SO<sub>2</sub> and NO<sub>x</sub> gases released during combustion.

#### a. Acidification

Acidification effects are related to increases in the acidity of water and soil in ecosystems. Increases in water acidity can impair the ability of certain types of fish and other biota to grow, reproduce, and hence, survive. In some acidified lakes and streams, entire populations of fish species have disappeared. For example, many lakes in the higher Adirondack mountains of New York and many streams in the Appalachian mountain region have experienced loss of trout and other biodiversity losses due to high acidity levels in the water (NAPAP, 1993:76). Increases in soil acidity can impair the ability of some types of trees to grow and resist disease. For example, growth reductions and injury to red spruce on high elevation ridges of the Appalachian mountains from Maine to Georgia have been linked to nutrient leaching caused by high soil acidity and deposition and primarily linked to a predisposition to frost damage from highly acidic cloud water (Johnson et al, 1992). The effects of acid deposition on forested ecosystems is an important research issue primarily because the observational data are inclusive (i.e., trees react very slowly to damaging influences).

# *i.* Lakes, Streams, and Watershed Ecosystems

Recent scientific studies indicate the amount of N that can be sequestered and retained in certain watersheds by biological processes is limited (US EPA, 1995:11). As these watersheds move towards N saturation, nitrate and, to a lesser extent, nitrite can begin to leach into surface waters, accelerating the process of long-term chronic acidification. Adding N to freshwater ecosystems that are rich in phosphorus can eutrophy as well as acidify the waters. Eutrophication also leads to decreased diversity of both plant and animal species (Vitousek et al, 1997:10).

Atmospheric deposition of N compounds plays a significant role in short-term episodic

acidification, which occurs when pulses of highly acidic water enter lakes and streams during storm flow, spring snowmelt or autumn rains after prolonged summer drought. Acidic episodes can expose aquatic organisms (e.g., fish, amphibians) to "acid pulses" containing high concentrations of inorganic monomeric aluminum (Al<sub>im</sub>), which is highly toxic to fish, often during the spawning season in the Spring. Episodic acidification can affect poorly buffered surface waters in many regions, including high elevation areas in the Mid-Atlantic and the West, as well as the Northeast (US EPA, 1995:14, 25).

The relative contributions of N and sulfur compounds, primarily  $NO_3^{-1}$  and  $SO_4^{-2-}$ , to the problem of surface water and soil acidification differs among regions and sites. The relative contributions depend not only on external differences in the deposition rates of these chemicals, but also on differences among the capacities of receptor watersheds to retain N and sulfur and to "buffer" against pH changes (i.e. alkalinity or hardness). Many areas in the West are more affected by N deposition, particularly dry deposition, than by sulfur deposition (US EPA, 1995:56).

Acidified watershed ecosystems can show signs of recovery following decreases in acid deposition rates. According to the Acid Deposition Standard Feasibility Study (US EPA, 1995), in watersheds where atmospheric deposition of sulfur has been and will continue to be decreased (commensurate with decreases in  $SO_2$  emissions under Title IV of the CAA), environmental modeling has projected a range of benefits (i.e., fewer acidic ecosystems) in sensitive ecosystems. The number of acidic systems are substantially fewer than the model projects without the  $SO_2$  emissions reductions in Title IV. Recovery rates depend primarily on the rates of pollutant decreases, ecosystem N retention processes, time lags caused by long-term biological process responses, and other possible changes in soil chemistry. Although watershed N saturation is widely accepted in the research community, it is also broadly recognized that there are uncertainties associated with the rate at which a watershed may become N saturated. However, additional  $NO_x$  emissions reductions would likely produce a two-fold benefit by decreasing acid deposition rates and lengthening the average time before watersheds reach N saturation.

### *ii.* Forests and Vegetation

Past assessments of the impacts of acid deposition on forests and vegetation have focused primarily on  $SO_2$  and sulfur deposition, largely because N is an essential nutrient for many biological processes (Atkinson,1993; Sommerville et al, 1989). Because N is commonly used as a fertilizer, it was thought that any atmospherically deposited N would be quickly and beneficially incorporated into plant and tree organisms (US EPA, 1995:11). Like aquatic ecosystems, the biological demand for N in forest ecosystems and other vegetation varies across geographical areas and by season. It is also highly dependent on factors such as tree/plant species (e.g., deciduous- species trees tend to have greater demand for N per unit biomass than coniferousspecies trees), soil type, forest age, prevalence of disease and other stresses such as extreme cold or drought, and land management practices (e.g., use of fertilizers, liming, or other cultivation methods) (US EPA, 1995:11). Acidification effects on health and productivity of forests and other vegetation are divided into two types: (1) direct effects on foliar organs and (2) indirect or soil-mediated effects resulting from acidification and physical/chemical alteration of the soil. Direct acidification effects might include foliar damage, erosion of leaf cuticle waxes, and changes in the physiology of tree leaves (Society of American Foresters, 1984). Soil-mediated acidification effects include toxic effects on roots as well as possible changes in nutrient availability, reproductive and regenerative processes.

Increasing evidence reveals that dry deposition is usually a significant portion of total atmospheric deposition (wet + cloud + dry) of both sulfur and N. For example, across all sites included in a recent review, dry deposition ranged from 9 to 59 percent of total deposition for sulfur (S), 25 to 70 percent for nitrate, and 2 to 33 percent for ammonia (Lovett, 1994; 629-650). Thus, in many areas N is taken up by foliage primarily in dry chemical form (e.g., as nitric acid vapor), rather than with deposition in precipitation. The response of forest ecosystems to direct effects of atmospheric deposition of both sulfur and N depend on the nature and timing of the deposition as well as the type of vegetation exposed. Some species appear less tolerant than others (i.e., spruce-fir ecosystems appear to be the most sensitive) and younger trees appear more vulnerable than mature trees.

Considerably more but still limited research has been performed on soil-mediated acidification effects since soils, together with climate, determine the productivity of terrestrial ecosystems. These studies have focused primarily on decreases in available base cation plant nutrients below amounts required for plant growth; and increased mobilization and availability of toxic aluminum (Al) and other metal ions (Brandt, 1993:14, 31). In certain soils, N deposition can deplete nutrients by leaching calcium (Ca), magnesium (Mg), and potassium (K). These important cations are often replaced by hydrogen ions (H<sup>+</sup>) which, together with increased mobilization of aluminum, can greatly increase soil acidification. Significant increases in sulfate and/or nitrate concentration will lead to preferential mobilization, availability, and toxicity of aluminum over base cations (e.g., Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>) in soils with low base saturation, such as the soils commonly found in high-elevation sites in the Northeast and Southeast (Turchenek et al., 1987; Turner et al., 1986). Increased concentrations and mobility of aluminum are linked with root damage and limited uptake of root calcium and magnesium. (Shortle and Smith, 1988).

The timing of aluminum concentration peaks is also important. Toxic aluminum peaks related to nitrate fluctuations commonly occur in late summer or early fall when soil temperatures and root growth are usually high (Joslin et al., 1992). It has been estimated that up to 3 percent of forested soils in the eastern United States could have toxic levels of trace elements in solution or could act as a source of high levels of acidity to surface waters, thus contributing to the acidification of watershed ecosystems discussed previously. Further, up to 40 percent of eastern soils may be sensitive to changes in nutrient status that could result in reduced forest growth or additional acidification of surface waters (Turner et al., 1986).

Forest ecosystems and other vegetated regions (e.g. crop and grasslands) are also susceptible to adverse excess N loading effects analogous to eutrophication in aquatic ecosystems.

These N loading effects result from deposited N of *all* forms (i.e., including forms other than acidic nitrate such as ammonia and dissolved organic N) and tend to occur when the demand by plants and heterotrophic soil organisms for N has been substantially satisfied (i.e., the ecosystem is approaching N saturation). N deposition to forest ecosystems can affect competitive relationships across tree/plant species and can therefore change species composition and/or diversity. Other potential adverse N loading effects include decreased uptake of nutrients from soil, increased susceptibility to insect and disease attack, and altered reproductive or regenerative processes (US EPA, 1991).

Evidence has accumulated suggesting N availability in certain forest ecosystems are in excess of plant and microbial demand. Early indicators of N saturation have implications to forest ecosystems over large geographic areas. Possible effects include elevated concentrations of nitrate, aluminum, and hydrogen in streams, which would decrease water quality, increase susceptibility to frost damage or other disruptions of physiological function that would lower productivity in certain forest types, increased cation [nutrient] leaching from soils and nitrate losses that would lead to lower soil fertility and increased acidity (Aber, et al., 1989). Additionally, recent research conducted in the Colorado Front Range demonstrates that high elevation (alpine and subalpine) ecosystems may be nearly N saturated at current levels of N deposition. The results suggest that the Colorado Front Range may be an early warning indicator of N saturation for other high-elevation catchments in the Rocky Mountains and the western United States and an indicator for disruption of N cycling in forested ecosystems at lower elevations as well. (Williams, et al., 1996)

Results of twelve years of experimental N addition to grassland plots in Minnesota have shown reductions in grassland biodiversity associated with N loadings. N added to research plots resulted in the loss of almost all native prairie grass species and to dominance by a weedy quackgrass. These results indicate that N loading can be a major threat to grassland ecosystems, causing loss of diversity, increased abundance of nonnative species, and the disruption of ecosystem functioning. (Wedin, et al., 1996)

Finally,  $NO_x$  is a primary  $O_3$  precursor and the damaging effects of  $O_3$  on forest ecosystems have been studied more comprehensively than those related to excess N loading and acidification.  $O_3$  is the most destructive pollutant in forest ecosystems (deSteiguer et al., 1990). The injurious effects of  $O_3$  on plants include visible damage to foliage, decreased growth of roots and shoots, decreased yield, changes in quality of harvest, and changes in susceptibility to other stresses. (US EPA, 1993).

# b. Materials and Structures

The role of atmospheric N deposition in metals corrosion has not been completely resolved; some suggest, on the basis of laboratory evidence, that  $NO_x$  appreciably increases the corrosive effect of SO<sub>2</sub> (NAPAP, 1993:93-94). It has been estimated that 31-78 percent of the dissolution of galvanized steel and copper is attributable to wet and dry acid deposition (NAPAP, 1993:93). Deposited acids corrode and dissolve the protective zinc coatings on these surfaces,

and as a result, the metal underneath rusts. The specific role of N-based acids in the process has not yet been established. In the late 1980s, NAPAP and the Economic Commission for Europe initiated several projects in the United States and Europe to clarify the scientific foundation linking acid deposition and materials damage (NAPAP, 1993:93). These projects include research to investigate the mechanisms by which N deposition directly impacts or works with other pollutants to damage structural and other materials.

Acid deposition also damages exterior paints applied to wood and metal substrates. Special paint formulations involving different organic and inorganic binders, pigments, and additives have been developed to resist corrosion and spotting from acid deposition (NAPAP, 1993:96). To maximize durability, these special finishes are applied under factory-controlled conditions. The costs to automotive manufacturers for including acid-resistant features have been estimated by the EPA and NAPAP to be as high as \$400 million annually (US EPA, 1995:97). Acid deposition can also accelerate the deterioration of stone through processes of erosion, solubilization, blackening of the stone surface, and cracking (US EPA, 1995:96-97). Acidic-related damage to cultural and historic buildings, monuments, and structures increases annual maintenance and reparation costs, which can be extensive. Thus, potentially large economic benefits could be associated with lessened physical materials damage achieved, in part, through additional  $NO_x$  emissions reductions.

# 4. How much reduction is needed?

Our current knowledge of the science of  $NO_x$  and acid deposition does not support quantitative assessments of the tons of  $NO_x$  needed beyond the CAA nationally or by region to protect sensitive aquatic and forest ecosystems or to reduce acidic-related damage to materials, structures, and cultural or historic resources. Nonetheless, model projections from EPA's recent Acid Deposition Standard Feasibility Study (October 1995) indicate that N deposition may play an important role in ongoing and future acidification of sensitive watershed ecosystems, and may equal or exceed the effects of sulfur deposition. The extent of potential future effects depends on how rapidly atmospheric N deposition moves watersheds toward a state of N saturation, i.e., where input of N exceeds biological uptake of N on an annual basis. The time to watershed N saturation will vary depending on forest age, historic and future rates of N deposition, future changes in ambient temperatures, water stress, land use as well as other variables.

The United States Congress directed EPA, in Section 404 (Acid Deposition Standards Study) of the CAA, to provide a report on the feasibility and effectiveness of an acid deposition standard or standards to protect sensitive and critically sensitive aquatic and terrestrial resources. The EPA's <u>Acid Deposition Standard Feasibility Study: Report to Congress</u> (US EPA, 1995) fulfills this requirement by integrating state-of-the-art ecological effects research, emissions and source receptor modeling, and evaluation of implementation and cost issues related to the feasibility of establishing and implementing an acid deposition standard or standards.

An acid deposition standard is a rate of deposition (most likely in units of kilograms of

pollutant per hectare<sup>2</sup> per year) that provides a predetermined amount of protection to specific ecological resources. Aquatic systems are the natural resources most at risk from acid deposition and those most amenable to quantitative assessment. Other ecological resources such as highelevation red spruce forests in the eastern United States and Canada may also be at risk, but less is known about the effects process, and the rate and extent of impacts on those resources. Target populations of Adirondack lakes, Mid-Atlantic streams, and Southern Blue Ridge streams were selected as case studies for detailed analysis in the Acid Deposition Standard Feasibility Study because they represent ecosystems that receive fairly high amounts of acid deposition, are sensitive to acid deposition, have the best historical data, and have been the focus of scientific studies. While many surface waters in western North America are as sensitive as, or more sensitive than, aquatic systems in the East, acid deposition rates in the West are currently sufficiently low that the risk of chronic (long-term) acidification to resources in the West is low and is expected to remain low for the next 50 years. Episodic acidification from spring snow melts, which adversely affects some eastern surface waters, also affects high elevation western surface waters. (US EPA, 1995:xiv).

For the Acid Deposition Standard Feasibility Study, EPA scientists modeled the potential combined effects of atmospheric deposition of both sulfur and N on the chemistry of acidsensitive lakes and streams in the regions selected for in-depth study: Adirondacks, Mid-Appalachian Region, and Southern Blue Ridge Province. Model simulations projected water chemistry responses out to the year 2040. Projections of sulfur and N deposition rates were based on results expected from implementation of the 1990 CAA amendments as well as other more restrictive deposition reduction scenarios using EPA's Regional Acid Deposition Model (RADM). The modeling incorporated a decision-model based estimate of SO<sub>2</sub> emission allowance trading and the Canadian SO<sub>2</sub> control program. Explicit watershed models and data to estimate the times required for watersheds to reach N saturation were unavailable at the time of the Study therefore, EPA scientists assumed an encompassing range of times (50 years, 100 years, 250 years, and never) to watershed N saturation and then estimated the potential consequent effects on surface water Acid Neutralizing Capacity (ANC). (ANC is a commonly used measure of the concentration of dissolved compounds [e.g., carbonate, bicarbonate, borates, and silicates] in fresh water which collectively tend to create less acidic conditions. Surface waters with higher ANC are generally more resistant to acidification.) This innovative modeling component of the Acid Deposition Standard Feasibility Study is referred to as the Nitrogen Bounding Study (NBS) in that, given the uncertainties associated with the time when a watershed may reach N saturation, the results effectively bounded the range of possible water chemistry outcomes. The NBS received external technical peer review and the entire Feasibility Study has been peer-reviewed by EPA's Science Advisory Board (US EPA, Appendix D, 1995).

Although model projections in the Acid Deposition Standard Feasibility Study are for three specific target populations, i.e., groups of lakes or streams with watersheds of similar size, land, and other characteristics, not even for all watersheds in the study regions, they signal a

 $<sup>^2</sup>$  A hectare is a unit of surface measure equal to 10,000 square meters.

direction of probable need for substantial additional reductions in year-round NO<sub>x</sub> emissions in the Eastern United States For example, it was estimated that a 40-50 percent decrease in SO<sub>2</sub> and NO<sub>x</sub> emissions beyond the CAA may be required to keep the number of chronically acidified lakes in the Adirondacks at 1984 proportions, if these watersheds move towards N saturation in 100 years<sup>3</sup> (US EPA, 1995:xvi). Without additional emissions reductions, the model projects the number of acidic lakes in the Adirondacks could increase by almost 40 percent by 2040, if these watersheds move towards N saturation in 100 years. As described, the modeling effort encompasses a range of responses based on time to N watershed saturation. For example, in the case in which saturation never occurs in the Adirondacks, the number of acidified lakes is lowered by 40 percent due to the SO<sub>2</sub> emissions reduction in the CAA. The effects on episodic acidification of lakes and streams would be even more pronounced as it is now understood that high nitrate levels are largely responsible for acidic episodes during snowmelt and high stream flow periods in the Northeast and probably high-elevation areas in other regions of the United States (Wigington et al., 1996; US EPA, 1995).

Recent results from the Bear Brook Watershed Manipulation Experiment illustrate the rapidity with which forested watersheds in the Northeast may reach N saturation in response to increased atmospheric N deposition (Scofield, 1995; Norton et al., 1994). :. Increased leaching of nitrate from forested catchments into streams or lakes could lead to increases in surface water acidification in some areas that could offset increases in ANC (i.e., reductions in acidity) expected from decreases in SO<sub>2</sub> emissions under the CAA.

5. How much reduction will be achieved with current and projected Title IV programs?

Under the current rule for the Acid Deposition  $NO_x$  Emission Reduction Program (40 CFR Part 76; FR 18751, April 13, 1995),  $NO_x$  emissions from existing coal-fired electric utility power plants will be decreased by over 400,000 tons per year between 1996 and 1999 (Phase I) and by over 1.5 million tons per year beginning in 2000 (Phase II). These decreases are achieved by 857 dry bottom wall-fired and tangentially fired boilers (Group 1). The annual cost of this regulation to the electric utility industry is estimated as \$267 million (in 1990 dollars), resulting in an overall cost-effectiveness of \$227 per ton of  $NO_x$  removed. The nationwide cost impact on electricity consumers is an average increase in electricity rates of approximately 0.21 percent, beginning in 2000 (61 FR 1442).

The Phase II Acid Deposition  $NO_x$  Emission Reduction Program will achieve an additional reduction of 890,000 tons of  $NO_x$  per year from existing coal-fired electric utility power plants beginning in 2000. One hundred twenty thousand (120,000) tons would come from lowering the

<sup>&</sup>lt;sup>3</sup> Observational data (currently being collected and analyzed by the New York State Department of Environmental Conservation) which compare the amount of nitrate falling on several Adirondack watersheds with the amount of nitrate leaving these watersheds in stream water indicate the watersheds may be nearing N saturation (Simonin, 1996; Evans et al., 1996).

emission limits for 580 Group 1 boilers affected in Phase II; 77 percent of these boilers are located in the Eastern United States, defined by the 37 states adjacent to and east of the Mississippi River. The additional tons would come from establishing emission limits for 190 high-emitting Group 2 boilers (i.e., cell burners, cyclones, wet bottom boilers, and vertically fired boilers); 89 percent of these boilers are located in the Eastern United States.

The Phase II Acid Deposition  $NO_x$  Emission Reduction Program appears to represent a singular regulatory opportunity for controlling high-emitting Group 2 boilers, which typically emit  $NO_x$  at rates in excess of 1.0 lb/mmBtu. The majority of these coal-fired boilers are located outside designated  $O_3$  nonattainment areas in the states of Illinois, Indiana, Kentucky, Michigan, Missouri, Ohio, and West Virginia. EPA modeling analyses show that transport of  $O_3$  and  $O_3$  precursors (primarily  $NO_x$ ) from upwind areas in the Eastern United States contributes significantly to  $O_3$  exceedances in virtually all nonattainment areas in the Northeast Ozone Transport Region (60 FR 45583). Further, simulations on EPA's Regional Acid Deposition Model (RADM) indicate not only that utility sources of N contribute the majority of deposits on the western side of the Chesapeake Bay, but also that the areal extent of the Chesapeake Bay airshed (which encompasses all or parts of Indiana, Kentucky, Ohio, and West Virginia as well as 10 other states) underestimates the areas contributing atmospheric sources of N deposition entering the Bay (Dennis, 1995).

The average cost-effectiveness of utility  $NO_x$  controls under the rule compares favorably to many of the other pollution control measures being considered by states to mitigate persistent O<sub>3</sub> nonattainment and/or N-based eutrophication water quality problems. For example, decreases in NO<sub>x</sub> emissions from coal-fired power plants are comparable or less expensive to implement than certain alternative controls for reducing N loadings to the Chesapeake Bay from area sources (farms, forests), even without counting the "clean air" benefits associated with the NO<sub>x</sub> emission reductions. Such alternatives, as well as others in the mobile source sector, are presently being considered by Maryland, Virginia, Pennsylvania, and the District of Columbia to achieve the 40 percent-decrease in controllable nutrient supplies to the Bay, to which these jurisdictions have committed. The average cost-effectiveness of these other controls are: chemical addition or biological removal of N from wastewater processing (\$4,000 to over \$20,000/ton N removed) and "management practices" to decrease N from fertilizers, animal waste, and other nonpoint sources (\$1,000 to over \$100,000/ton of N removed) (Camacho, 1993; Shuyler, 1992). While it is recognized that to address the Bay's excessive nutrient loading problem in the most efficient manner requires pursuing an integrated strategy of air, water, and agricultural pollution control practices, these relative cost-effectiveness ratios and modeling analyses suggest the additional NO<sub>x</sub> emissions reductions from coal-fired power plants in the acid deposition rule are a critical component of this strategy.

6. Summary

The primary goal of the Acid Deposition  $NO_x$  Emission Reduction Program is to reduce the multiple adverse effects of  $NO_x$ , a principal acid deposition precursor that contributes to air and water pollution, by substantially decreasing annual emissions from existing coal-fired power plants. Acid deposition occurs when airborne acidic and acidifying compounds, principally sulfates  $(SO_4^{2^-})$  and nitrates  $(NO_3^{-})$ , which can be transported over long distances, return to the earth through rain or snow ("wet deposition"), through fog or cloud water ("cloud deposition"), or through transfer of gases or particles ("dry deposition"). According to section 401(a)(1) of the CAA, acid deposition "represents a threat to natural resources, ecosystems, visibility, materials, and public health." Since  $NO_x$  emissions from the burning of fossil fuels at power plants also contribute to the formation of ground-level  $O_3$  and nitrate PM in the air, ambient levels of  $NO_2$ , and excessive N loadings to the Chesapeake Bay and other estuaries, decreases in  $NO_x$  emissions under the Acid Deposition Program are expected to have multiple and synergistic beneficial impacts on public health and welfare.

The Acid Deposition  $NO_x$  Emission Reduction Program consists of a two-stage program which, analogous to the Acid Deposition allowance program for  $SO_2$  emission reduction, is implemented in two phases. The first stage of the program, authorized by section 407(b)(1) of the CAA and implemented pursuant to 40 CFR Part 76, promulgated in April 1995, will decrease annual  $NO_x$  emissions by over 400,000 tons per year, beginning in 1996, from 170 Phase I affected units<sup>4</sup> with Group 1 boilers (i.e., dry bottom wall-fired boilers and tangentially fired boilers). An additional  $NO_x$  emissions reduction of over 200,000 tons per year will probably be realized, beginning in 1997, from about 250 "early election" Phase II units with Group 1 boilers which voluntarily opted into the Phase I program. The total  $NO_x$  emissions reduction that would be achieved by applying LNB technology under the April 13, 1995 rule is estimated at about 1.2 million tons per year, beginning in 2000.

In December 1996, EPA promulgated regulations for implementing the second stage of the program, authorized by section 407(b)(2) of the CAA. Compliance with the rule would achieve an additional NO<sub>x</sub> emissions reduction of 890,000 tons per year, beginning in 2000, from existing coal-fired units affected in Phase II. Seventy-seven percent of the Group 1, Phase II boilers and 89 percent of the Group 2 boilers are located in states adjacent to or east of the Mississippi River. EPA modeling analyses show that utility sources of N in this 37-state region contribute significantly to the acidification of certain watershed ecosystems (US EPA, 1995), excess N deposits to the Chesapeake Bay (Dennis, 1995), and O<sub>3</sub> exceedances in virtually all nonattainment areas in the densely populated Northeast Ozone Transport Region (60 FR 45583). The total NO<sub>x</sub> emissions decrease under the statutory authority of Title IV (Acid Deposition Control) is estimated at about 2 million tons per year, beginning in 2000.

Such emissions decreases may not be adequate, however, to protect sensitive watershed ecosystems in the Northeast and Mid-Atlantic regions as well as in high-elevation areas in the West and other regions. Recent model projections from EPA's <u>Acid Deposition Standard</u> <u>Feasibility Study: Report to Congress</u> (US EPA, 1995) signal a direction of probable need for

<sup>&</sup>lt;sup>4</sup> Known as Table 1 units. These 170 units represent the coal-fired units with Group 1 boilers that are listed in Table 1 of 40 CFR 73.10 (a) of the Acid Rain Program Regulations and are subject to 40 CFR Part 76, Acid Rain  $NO_x$  Emission Reduction Rule.

substantial additional reductions in year-round NO<sub>x</sub> emissions. For example, it was estimated that a 40-50 percent decrease in SO<sub>2</sub> and NO<sub>x</sub> emissions beyond the CAA may be required to keep the number of chronically acidified lakes in the Adirondacks at 1984 proportions, if the time to N saturation in these watersheds is 100 years or less. Without additional emissions reductions, the model projects the number of acidic lakes in the Adirondacks could increase by almost 40 percent by 2040, assuming N saturation in 100 years. There are uncertainties associated with determining the rate at which a watershed may reach N saturation and therefore the EPA's Study provides a range of possible responses. However, the magnitude and direction of projected responses point towards a need for further emissions reductions to protect sensitive ecosystems. The negative effects of no additional emissions reductions could be even more pronounced on episodic acidification of lakes and streams in the Northeast (and potentially high-elevation areas in other regions) where high nitrate levels are largely responsible for acidic episodes during snowmelt and high stream flow periods (Wigington et al., 1996; US EPA, 1995). Thus, wintertime NO<sub>x</sub> emissions reductions are especially important to lessening the incidence and severity of acidic episodes in certain areas. Continuous year-round NO<sub>x</sub> controls appear to be the most beneficial for decreasing acid deposition damage to natural resources.

#### References

Aber, John D., Knute J. Nadelhoffer, Paul Steudler, and Jerry M. Melillo. 1989. "Nitrogen Saturation in Northern Forest Ecosystems." *BioScience* Vol. 39 No. 6. June.

Atkinson, Giles. 1993. "The Impact of Acid Rain on Crops." Reprint from Center for Social and Economic Research on the Global Environment, University College London and University of East Anglia, UK.

Brandt, C. Jeffrey. 1994. Acidic Deposition and Forest Soils: Potential Changes in Nutrient Cycles and Effects on Tree Growth. Battelle Pacific Northwest Laboratory. EPA 600/R-94/153 ERL-COR 817. May.

Camacho, R. 1993. "Financial Cost-Effectiveness of Point and Nonpoint Source Nutrient Reduction Technologies in the Chesapeake Bay Basin." Report No. 8 of the Chesapeake Bay Program Nutrient Reduction Strategy Reevaluation. Washington, DC. U.S. Environmental Protection Agency, February.

Church, M.R. 1997. "Hydrochemistry of forested catchments." Annual Reviews of Earth and Planetary Sciences. Vol. 25 (preprint).

de Steiguer, J.E., J.M. Pye, and C.S. Love. 1990. "Air pollution damage to U.S. forests." *Journal of Forestry*, Vol. 88, No. 8, p. 17. August.

Evans, C.D., T.D. Davies, P.J. Wigington, M. Tranter, and W.A. Kreiser. 1996 (in press). "Use of factor analysis to investigate processes controlling the chemical composition of four streams in the Adirondack mountains," New York. *Journal of Hydrology*.

*Federal Register*. 1995. 40 CFR Part 76. "Acid Rain Program: Nitrogen Oxides Emission Reduction Program." Vol. 60, No. 71, pp. 18751. Thursday, April 13, 1995. Direct final rule.

*Federal Register*. 1995. 40 CFR, Parts 80, 86 and 89. "Control of Air Pollution From Heavy-Duty Engines." Vol. 60, No. 169, pp. 45580. Thursday, August 31, 1995. Proposed rule.

*Federal Register.* 1996. 40 CFR Part 76. "Acid Rain Program: Nitrogen Oxides Emission Reduction Program." Vol. 61, No. 13, pp. 1442. Friday, January 16, 1996. Proposed rule .

Johnson, A.H., S.B. McLaughlin, M.B. Adams, E.R. Cook, D.H. DeHayes, C. Eager, I.J. Fernandez, D.W. Johnson, R.J. Kohut, V.A. Mohnen, NM.S. Nicholas, D.R. Peart, G, A. Schier, and P.S. White. 1992. "Synthesis and Conclusions from Epidemiological and Mechanistic Studies of Red Spruce Decline." Pp. 385-412. In C. Eager and M.B. Adams, eds. *Ecology and Decline of Red Spruce in the Eastern United States*. Springer Verlag, New York.

Joslin, J.D., J.M. Kelly, and H. Van Meigroet. 1992. "Soil chemistry and nutrition of North American spruce-fir stands: evidence for recent change." *J. Environ. Qual.* 21:12-30.

Lovett, G.M. 1994. "Atmospheric deposition and pollutants in North America: an ecological perspective." *Environmental Applications* 4:629-650.

National Acid Precipitation Assessment Program (NAPAP). 1990. Integrated Assessment . External Review Draft.

"National Acid Precipitation Assessment Program (NAPAP)." 1992 Report to Congress. June 1993.

Norton, S.A., J.S. Kahl, I.J. Fernandez, L.E. Rustad, J.P. Scofield and T.A. Haines. 1994. "Response of the West Bear Brook Watershed, Maine, USA, to the addition of  $(NH_4)_2SO_4$ : 3-year results." *Forest Ecology and Management*. 68:61-73.

Paerl, H.W. 1993. "The Emerging Role of Atmospheric Nitrogen Deposition in Coastal Eutrophication: A Biogeochemical and Trophic Perspective." *Canadian Journal of Fisheries and Aquatic Sciences*. 50:2254-2269.

Scofield, J.P. 1995. "Annual Progress Report for Cooperative Agreement CR-816261. Watershed Manipulation Project: Maine Site Group."

Shortle, W.C. and K.T. Smith. 1988. "Aluminum-induced calcium deficiency syndrome in declining red spruce." *Science* 240:1017-1018.

Shuyler, L.R. 1992. "Cost Analysis for Nonpoint Source Control Categories in the Chesapeake Basin." Annapolis, MD: U.S. Environmental Protection Agency . Chesapeake Bay Program.

March.

Simonin, H.A. 1996. Unpublished data.

Society of American Foresters. 1984. "Acidic Deposition and Forests." Bethesda, MD.

Sommerville, M.C., S.E. Spruill, J.O. Rawling, and V.M. Lesser. 1989. "Impact of Ozone and Sulfur Dioxide on the Yield of Agricultural Crops." Technical Bulletin 292. North Carolina Agricultural Research Service, North Carolina State University. November.

Taylor, G.E., D.W. Johnson, and C.P. Anderson. 1994. "Air pollution and forest ecosystems: a regional to global perspective." *Ecological Applications*. 4:662-689.

Turchenek, L.W., S.A. Abboud, C.J. Tomas, R.J. Fessenden, and N. Holowaychuck. 1987. "Effects of acid deposition on soils in Alberta." Acid Deposition Research Program, Calgary, Alberta, Canada.

Turner, R.S., R.J. Olson, and C.C. Brandt. 1986. "Areas having soils characteristics that may indicate sensitivity to acidic deposition under alternative forest damage hypotheses." ORNL/TM-9917. Environmental Sciences Division. Pub. No. 2720. Oak Ridge National Laboratory .

US EPA. 1991. "Air Quality Criteria for Oxides of Nitrogen: External Review Draft," EPA 600/8-91/049bA, Environmental Criteria and Assessment Office, Office of Health and Environmental Assessment, Research Triangle Park, NC. August.

US EPA. 1993. Air Quality Criteria for Ozone and Related Photochemical Oxidants. Volume II of III. EPA 600/Ap-93/004d. April.

US EPA. 1995. Acid Deposition Standard Feasibility Study: Report to Congress. EPA 430-R-95-001a. October.

Vitousek, P.M., J. Aber, R.W. Howarth, G.E. Likens, P.A. Matson, D.W. Schindler, W.H. Schlesinger, and G.D. Tilman, "Human Alteration of the Global Nitrogen Cycle: Causes and Consequences," *Issues in Ecology*, Number 1, Spring 1997.

Wedin, David A. and David Tilman. 1996. "Influence of Nitrogen Loading and Species Composition on the Carbon Balance of Grasslands." *Science*, Vol 274, December 6, 1996.

Wigington, J.P, D.R. DeWalle, P.S. Murdoch, W.A. Kretser, H.A. Simonin, J. Van Sickle, and J.P. Baker. 1996 (in press). "Episodic acidification of small streams in the Northeastern United States II. Ionic controls of episodes." *Ecological Applications* 6(2).

Williams, Mark W., Jill S. Baron, Nel Caine, Richard Sommerfield and Robert Sanford, Jr. 1996.

"Nitrogen Saturation in the Rocky Mountains." *Environmental Science & Technology* Vol 30 No 2.

- B. Nitrogen Dioxide
- 1. Goal of the Program

The EPA has established national ambient air quality standards (NAAQS) for nitrogen dioxide (NO<sub>2</sub>) designed to protect public health and welfare. The goal of the program is to first achieve these clean air standards throughout the country and then to maintain the standards. Control of NO<sub>x</sub> emissions is needed locally in some areas to continue to maintain the NO<sub>2</sub> NAAQS.

2. Status of the Program

Section 109 of the CAA directs the EPA Administrator to propose and promulgate primary and secondary NAAQS for pollutants identified under section 108. Section 109 defines a primary standard as that necessary to protect the public health, allowing an adequate margin of safety. A secondary standard, as defined in section 109, must specify an air quality concentration needed to protect the public welfare from any known or anticipated adverse effects associated with the presence of the pollutant in the ambient air. Welfare effects, as defined in section 302(h) of the CAA include, but are not limited to, effects on soils, water, crops, vegetation, materials, animals, wildlife, weather, visibility and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being.

States are primarily responsible for ensuring attainment and maintenance of the NAAQS. Under title I of the CAA, States are to submit, for EPA approval, State implementation plans (SIPs) that provide for the attainment and maintenance of such standards through control programs directed to sources of the pollutants involved. In addition, Federal programs provide for nationwide reductions in emissions of air pollutants through, for example, the Federal Motor Vehicle Control Program, which involves controls for automobile, truck, bus, motorcycle, and aircraft emissions.

The primary and secondary NAAQS for NO<sub>2</sub> is 0.053 parts per million (ppm) (100 micrograms per meter cubed) annual arithmetic average. In selecting the concentration for the current standard, the Administrator made judgments regarding the lowest concentrations at which effects were observed, sensitive populations, nature and severity of public health effects, and margin of safety. After assessing the evidence, the Administrator concluded that the annual standard of 0.053 ppm adequately protected against adverse health effects associated with long-term exposures and provided some measure of protection against possible short-term health effects. The June 19, 1985 Federal Register notice (50 FR 25532) provides a detailed discussion of the bases for the existing standard.

Currently, all areas of the United States are in attainment of the annual NO<sub>2</sub> NAAQS of 0.053 ppm (EPA, 1994). Los Angeles is the only city in the United States to record violations of the annual average NO<sub>2</sub> NAAQS during the past decade. In 1992, Los Angeles reported air quality measurements which met the NO<sub>2</sub> NAAQS for the first time.

In accordance with the provisions of sections 108 and 109 of the CAA, as amended, the EPA conducted a review of the criteria upon which the existing NAAQS for NO<sub>2</sub> are based. The revised criteria were published simultaneously with the issuance of the October 11, 1995 Federal Register notice of proposed rulemaking on the NO<sub>2</sub> NAAQS (60 FR 52874). After evaluating the revised health and welfare criteria under section 109(d)(1) of the Act, and considering public comment, the Administrator published a final rulemaking notice on October 8, 1996 (61 FR 52852) which concludes that it is not appropriate to propose any revisions to the primary and secondary NAAQS for NO<sub>2</sub> at this time. As described in the proposed and final rulemaking notices, EPA determined that a 0.053 ppm annual standard would keep annual NO<sub>2</sub> concentrations considerably below the long-term levels for which serious chronic effects have been observed in animals. Retaining the existing standard also provides protection against short-term peak NO<sub>2</sub> concentrations at the levels associated with mild changes in pulmonary function and airway responsiveness observed in controlled human studies.

3. Science of NO<sub>2</sub>

 $NO_2$ 

 $NO_2$  is a brownish, highly reactive gas that is formed in the ambient air through the oxidation of NO. As described in the "Ozone" section, emissions of NO play a major role in the formation of  $O_3$  in the lower atmosphere through a complex series of reactions with VOCs. Emissions of NO are rapidly oxidized in the atmosphere to  $NO_2$ .  $NO_x$  refers to the sum of  $NO_2$  and NO. Sources of NO ( $NO_x$ ) emissions are described in the "Introduction/Overview" section and Appendix C. The major sources of anthropogenic  $NO_x$  emissions are on-road vehicles and electric utilities.

#### Health Effects of Concern.

Based on the health effects information contained in EPA's Criteria Document (EPA, 1993), which evaluates key studies published through early 1993 and the Staff Paper (EPA, 1995), EPA has concluded that  $NO_2$  is the only nitrogen oxide sufficiently widespread and commonly found in ambient air at high enough concentrations to be a matter of public health concern. Exposure to  $NO_2$  is associated with a variety of acute and chronic health effects. Two general groups in the population may be more susceptible to the effects of  $NO_2$  exposure than other individuals. These groups include persons with pre-existing respiratory disease and children 5 to 12 years old. Individuals in these groups appear to be affected by lower levels of  $NO_2$  than individuals in the rest of the population. The health effects of most concern at ambient or near-ambient concentrations of  $NO_2$  include mild changes in airway responsiveness and pulmonary function in individuals with pre-existing respiratory illnesses and increases in respiratory illnesses in children (5-12 years old).

Regarding a short-term NO<sub>2</sub> standard, EPA concluded that, while short-term effects from  $NO_2$  are documented in the scientific literature, the available information is insufficient to provide an adequate scientific basis for establishing any specific short-term standard. However, the EPA has analyzed the relationship between short-term exceedances of NO<sub>2</sub> concentrations and the annual NO<sub>2</sub> mean to determine whether the annual standard would be protective of the acute effects being observed. In 1994, EPA analyzed air quality data from the period 1988-1992 to determine the estimated number of exceedances of various NO<sub>2</sub> short-term air quality indicators which would occur given attainment of a range of annual averages. The annual averages analyzed ranged from 0.02 to 0.06 ppm and included the current NO<sub>2</sub> NAAQS of 0.053 ppm. The 1-hour and daily concentration levels chosen for analyses were 0.15, 0.20, 0.25, and 0.30 ppm. The results of this analysis are reported in "Analysis of High 1 Hr NO<sub>2</sub> Values and Associated Annual Averages Using 1988-1992 Data" (McCurdy, 1994). It was concluded that areas attaining the current annual NO<sub>2</sub> NAAQS reported few, if any, 1 hour or daily exceedances above 0.15 ppm. Based on the analyses of this air quality data, it was concluded that the existing annual standard provides adequate protection against potential changes in pulmonary function or airway responsiveness (which most experts would characterize as mild responses occurring in the range of 0.2 to 0.5 ppm  $NO_2$ ). The adequacy of the existing annual standard to protect against potential pulmonary effects is further supported by the absence of documented effects in some studies at higher (3 to 4 ppm) concentrations of NO<sub>2</sub> (EPA, 1995, p. 43).

#### Welfare Effects Associated with Exposure to NO<sub>2</sub>

NO<sub>2</sub> and other N compounds have been associated with a wide range of effects on public welfare. The principal effects associated with N deposition include acidification and eutrophication of aquatic systems. Both processes can sufficiently lower water quality making it unfit as a habitat for most aquatic organisms and/or human consumption. Acidification of lakes from N deposition may also increase leaching and methylation of mercury in aquatic systems. Atmospheric N can enter aquatic systems either as direct deposition to water surfaces or as N deposition to the watershed.

The principal effects on soils and vegetation associated with excess N inputs include: (1) Soil acidification and mobilization of aluminum, (2) increase in plant susceptibility to natural stresses, and (3)modification of inter-plant competition. Atmospheric deposition of N can accelerate the acidification of soils and increase aluminum mobilization if the total supply of N to the system (including deposition and internal supply) exceeds plant and microbial demand.

# 4. How much reduction is needed to *maintain* the current standard?

As noted above, all areas of the United States are currently in attainment of the annual NO<sub>2</sub> NAAQS of 0.053 ppm. Los Angeles, the last city in the United States to record violations of the annual average NO<sub>2</sub> NAAQS, has reported air quality measurements which met the NO<sub>2</sub> NAAQS since 1992. The Los Angeles and New York areas generally have recorded the highest annual NO<sub>2</sub> ambient concentrations in the nation. These two areas are expected to decrease NO<sub>x</sub> emissions in the future to meet the O<sub>3</sub> and/or PM standards. The November 1994 SIP submittal

for the Los Angeles area includes a 59 percent decrease in  $NO_x$  emissions. The New York area will benefit from significant  $NO_x$  emissions reductions throughout the Northeast Ozone Transport Region through implementation of the September 1994  $NO_x$  Memorandum of Understanding. Also helpful in maintaining the  $NO_2$  standard are the broad scale  $NO_x$  emissions reductions from implementation of the acid deposition requirements.

Given the implementation of  $NO_x$  emissions reductions needed to achieve the various goals of the CAA, it appears that the  $NO_2$  standard will continue to be attained throughout the nation in the foreseeable future. Additional local and regional  $NO_x$  emissions decreases in areas with relatively high  $NO_2$  concentrations are planned to meet the  $O_3$  and PM NAAQS. Thus, implementation of  $NO_x$  emissions reductions for  $O_3$ , PM, and acid deposition is likely to assure maintenance of the  $NO_2$  standard.

5. How much reduction will be achieved with current and projected programs?

As described in Appendices A and B, substantial decreases in  $NO_x$  emissions will be achieved through implementation of several on-going CAA programs. These decreases will benefit the  $NO_2$  program.

6. Summary

Exposure to NO<sub>2</sub> is associated with a variety of acute and chronic health effects. The health effects of most concern at ambient or near-ambient concentrations of NO<sub>2</sub> include changes in airway responsiveness and pulmonary function in individuals with pre-existing respiratory illnesses and increases in respiratory illnesses in children. Currently, all areas of the United States are in attainment of the annual NO<sub>2</sub> NAAQS of 0.053 ppm. Through implementation of NO<sub>x</sub> emissions reductions related to acid deposition and attainment of the O<sub>3</sub> and PM NAAQS, it is likely that the NO<sub>2</sub> standard will continue to be attained throughout the nation in the foreseeable future.

#### References

McCurdy, T. R., "Analysis of high 1 hour NO<sub>2</sub> values and associated annual averages using 1988-1992 data," Report of the Office of Air Quality Planning and Standards, Durham, NC., 1994.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, revised criteria document, *Air Quality Criteria for Oxides of Nitrogen*, three volumes, EPA-600/8-91/049aF-cF, August 1993.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, final revised OAQPS Staff Paper, *Review of the National Ambient Air Quality Standards for Nitrogen Oxides: Assessment of Scientific and Technical Information*, EPA-452/R-95-005, September 1995.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *National Air Quality and Emissions Trends Report, 1993*, EPA-454/R-94-026, October 1994.

#### C. Ozone

#### 1. Goals of the Program

The EPA has established health and welfare standards for ground level<sup>5</sup>  $O_3$ , which is the major component of summertime "smog." The goal of the program is to achieve and maintain these clean air standards throughout the country. As described below, reactions of the emissions of NO<sub>x</sub> and VOCs result in the formation of  $O_3$  which can adversely affect public health and welfare in many areas. Decreases in NO<sub>x</sub> emissions may be needed locally in some areas to attain the  $O_3$  NAAQS. In other areas, regional scale NO<sub>x</sub> emissions reductions may be needed to help attain the  $O_3$  NAAQS in some downwind areas and/or to help maintain  $O_3$  levels below the standard in some attainment areas.

2. Status of the Program

#### The NAAQS

Section 109 of the CAA directs the EPA Administrator to propose and promulgate primary and secondary NAAQS for pollutants identified under section 108. Section 109 defines a primary standard as that necessary to protect the public health, allowing an adequate margin of safety. A secondary standard, as defined in section 109, must specify an air quality concentration needed to protect the public welfare from any known or anticipated adverse effects associated with the presence of the pollutant in the ambient air. Welfare effects, as defined in section 302(h) of the CAA include, but are not limited to, effects on soils, water, crops, vegetation, materials, animals, wildlife, weather, visibility and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being.

States are primarily responsible for ensuring attainment and maintenance of the NAAQS. Under title I of the CAA, States are to submit, for EPA approval, State implementation plans (SIPs) that provide for the attainment and maintenance of such standards through control programs directed to sources of the pollutants involved. In addition, Federal programs provide for nationwide reductions in emissions of air pollutants through, for example, the Federal Motor Vehicle Control Program under title II of the Act, which involves controls for automobile, truck, bus, motorcycle, and aircraft emissions.

#### History of NAAQS Reviews

<sup>&</sup>lt;sup>5</sup>Ground level (tropospheric)  $O_3$  refers to  $O_3$  occurring from the ground level through about 15 kilometers; stratospheric  $O_3$ , which occurs between about 15-50 kilometers altitude is discussed in section III.D of this document.

# Establishment of NAAQS for Photochemical Oxidants

On April 30, 1971, the EPA promulgated NAAQS for photochemical oxidants under section 109 of the Act (36 FR 8186). Identical primary and secondary NAAQS were set at an hourly average of 80 parts per billion (ppb) total photochemical oxidants not to be exceeded more than 1 hour per year.

#### Review and Revision of NAAQS for Photochemical Oxidants

EPA published proposed revisions to the original NAAQS in 1978 (43 FR 16962) and final revisions in 1979 (44 FR 8202). The primary standard was revised from 80 to 120 ppb; the secondary standard was set identical to the primary standard; the chemical designation of the standards was changed from photochemical oxidants to  $O_3$ ; and the form of the standards was revised from a deterministic form to a statistical form, which defined attainment of the standards as occurring when the expected number of days per calendar year with maximum hourly average concentrations greater than 120 ppb is equal to or less than one.

#### Subsequent Review of O<sub>3</sub> NAAQS

In 1982 (47 FR 11561), the EPA announced plans to revise the 1978 Criteria Document. On August 10, 1992 (57 FR 35542), the EPA published a proposed decision under section 109(d)(1) that revisions to the existing primary and secondary standards were not appropriate at that time. On March 9, 1993 (58 FR 13008), the EPA published a final decision concluding that revisions to the current primary and secondary NAAQS for O<sub>3</sub> were not appropriate at that time. Given the potential importance of new studies and the EPA's continuing concern about the health and welfare effects of O<sub>3</sub>, the March 9, 1993 notice emphasized the Administrator's intention to complete the next review of the NAAQS as rapidly as possible and, if appropriate, to propose revisions of the standards at the earliest possible date.

#### Most Recent Review of O<sub>3</sub> NAAQS

A series of peer-review workshops was held on draft chapters of the revised Criteria Document in July and September 1993, and a first external review draft was made available for the Clean Air Scientific Advisory Committee<sup>6</sup> and public review on January 31, 1994. The EPA review includes analysis of the following alternative primary standards: the current 1-hr standard of 120 ppb, with a maximum expected exceedance rate of one per year (averaged over 3 years); an 8-hr standard in the range of 70-90 ppb, with a maximum expected exceedance rate of five per year. Further information on this subject was published in the advance notice of proposed rulemaking on "National Ambient Air Quality Standards for Ozone and Particulate Matter" published in the June 12, 1996 Federal Register.

On December 13, 1996, EPA proposed in the Federal Register to change the O<sub>3</sub>

<sup>&</sup>lt;sup>6</sup> A standing committee of EPA's Science Advisory Board.

standards. The proposed revised standards would provide protection for children and other atrisk populations against a wide range of  $O_3$  induced health effects. As described in detail in that notice, EPA proposed to change the current standard in several respects: (1) attainment would be based on 8-hour averages of  $O_3$ , not 1-hour averages; (2) the acceptable concentration would be lowered from 120 ppb  $O_3$  to 80 ppb.

EPA then conducted an extensive public comment process, receiving approximately 57,000 comments at public hearings held across the country and through written, telephone and computer messages on the  $O_3$  and particulate standards proposal. The proposed standards were also subjected to an intensive inter-agency review process. A court order required EPA to finalize a PM standard by mid-July of this year, and EPA committed to a court to do the same for  $O_3$ .

The final air quality standards for  $O_3$  and PM were published in the <u>Federal Register</u> of July 18, 1997 (62 FR 38856). The  $O_3$  standards are the same as those proposed in 1996, with one significant change: the final standard is set at the average fourth highest concentration instead of the third; this should provide greater stability in the standard for businesses and communities by requiring more "bad air" days before an area is found to be out of attainment.

It should be noted that, with EPA's July 18, 1997 adoption of the new  $O_3$  and PM standards, greater emphasis might be needed on regional-scale  $NO_x$  emissions reductions to reach attainment of the new standard(s). The new standards result in more areas and larger areas with monitoring data indicating nonattainment (Figure II-1).

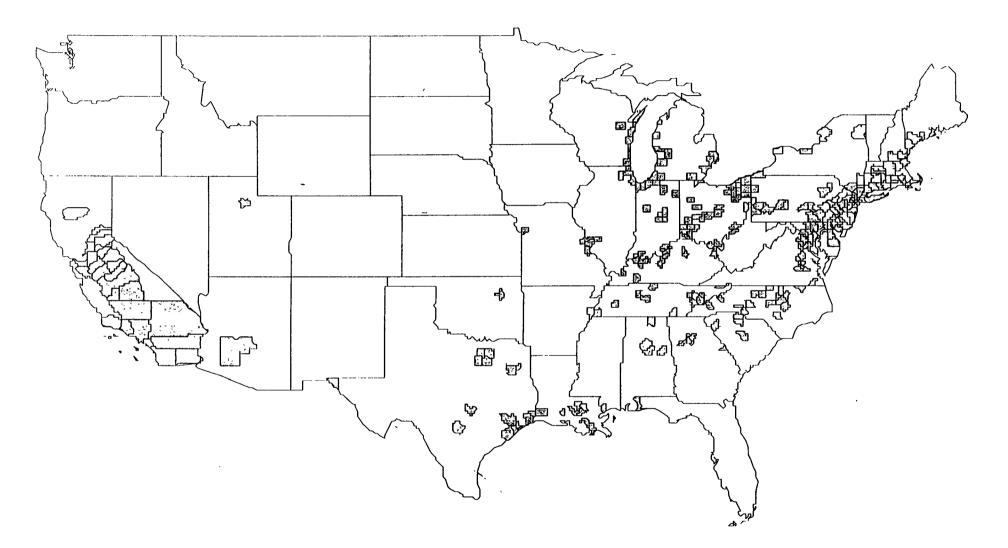
#### O<sub>3</sub> Nonattainment Areas

There are over 700 sites maintained by the States and local air pollution control agencies that measure ground level hourly  $O_3$  concentrations (EPA, 1995). Most of these monitoring sites are located in urban and suburban area locations, with far less density of sites in rural areas. Peak  $O_3$  concentrations typically occur during hot, dry, stagnant summertime conditions. Year-to-year meteorological fluctuations and long-term trends in the frequency and magnitude of peak  $O_3$  concentrations have a significant influence on an area's compliance status.

In 1991 EPA designated areas attainment and nonattainment for the 1-hour  $O_3$  standard (November 6, 1991 <u>Federal Register</u> at 56 FR 56694). At that time 98 areas were designated as not in attainment of the NAAQS for  $O_3$  (not including transitional and incomplete/no data areas). Over the last several years, many of these areas achieved the NAAQS for  $O_3$  and were redesignated to attainment, leaving a total of 60  $O_3$  nonattainment areas (as of July 28, 1997) shown in Figure II-2. More than one hundred million people live in areas designated as not attaining the 1-hour  $O_3$  standard.

# **Counties Potentially not Meeting Ozone Standard**

(0.08 ppm, 8-hour, 4th Maximum Concentration)



This map is based on measured air quality data from 1993 to 1995. When designations of nonattainment are made in two to three years they will be made on the most recent three years of quality assured data available for each area.

# **Classified Ozone Nonattainment Areas**

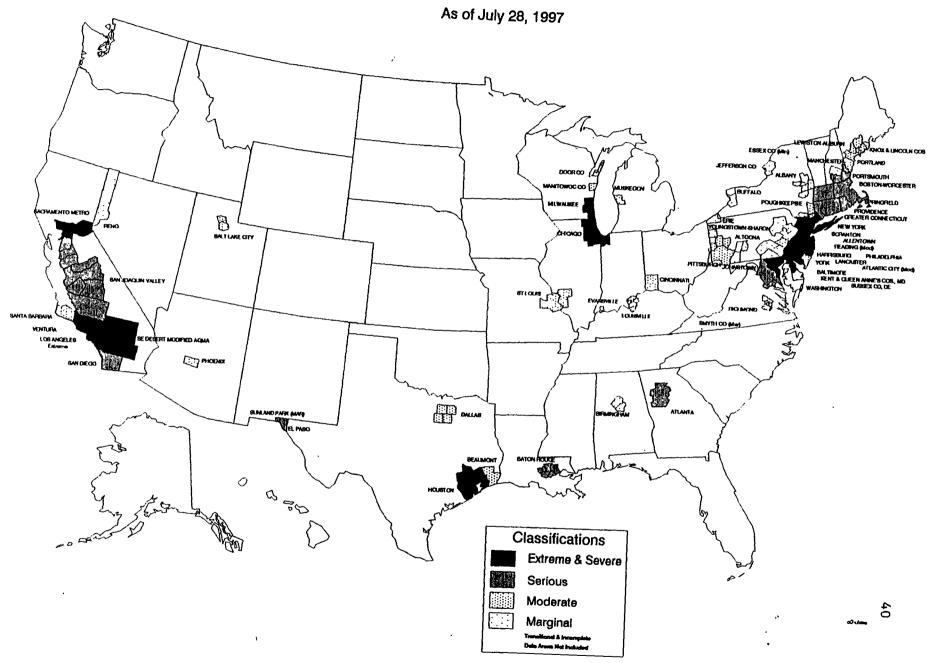


Figure II-2

# 3. Science of $NO_x$ and $O_3$

#### Health and Welfare Effects

In the lower levels of the troposphere  $O_3$  can have adverse effects. Concentrations of  $O_3$  near the earth's surface can result in damaging effects on human health, agricultural crops, ornamental plants, forests, and materials. A summary of these effects is provided below; for further information, see EPA's December 13, 1997 notice of proposed rulemaking and references cited therein, such as "Review of National Ambient Air Quality Standards for Ozone, Assessment of Scientific and Technical Information," OAQPS Staff Paper, June 1996.

A wide array of health effects has been attributed to short-term (1 to 3 hrs), prolonged (6 to 8 hrs), and long-term (months to years) exposures to elevated  $O_3$  levels. Those acute health effects induced by short-term exposures to  $O_3$  as low as 120 ppb, generally while engaged in heavy exercise, such as running, include: transient pulmonary function responses, transient respiratory symptoms and effects on exercise performance, increased airway responsiveness, transient pulmonary inflammation, increased susceptibility to respiratory infection, and increased hospital admissions and emergency room visits. Similar health effects have been observed following prolonged exposures to  $O_3$  as low as 80 ppb and at lower levels of exercise than for short-term exposures.

Welfare effects addressed by a secondary  $O_3$  NAAQS include, but are not limited to, effects on soils, water, crops, vegetation, man-made materials, animals, wildlife, weather, visibility and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being. Of these welfare effects categories, the effects of  $O_3$  on crops and vegetation are of most concern at concentrations typically occurring in the United States. By affecting crops and vegetation,  $O_3$  also directly and indirectly affects soils, water, animals, wildlife, and economic values, as well as aesthetic values, genetic resources, and natural ecosystems. Thus, providing protection for crops and vegetation would increase the protection afforded to these other related public welfare categories.

#### $O_3$ Background

 $O_3$  is a naturally occurring, trace constituent of the atmosphere.  $O_3$  concentrations vary by altitude, geographic location, and time. The natural component originates from three sources: (1) stratospheric  $O_3$  which is transported down to the troposphere, (2)  $O_3$  formed from the photochemically-initiated oxidation of biogenic and geogenic methane and carbon monoxide (CO) with biogenic  $NO_x$ , and (3)  $O_3$  formed from the photochemically-initiated oxidation of biogenic VOCs with biogenic  $NO_x$ . Lightning and soils are the major biogenic sources of  $NO_x$  emissions and play an important role in the oxidation of methane, carbon monoxide, and biogenic VOCs, though the magnitude of this natural part cannot be precisely determined (EPA, 1996). In remote locations--i.e., areas thought to be unaffected by anthropogenic sources-- $O_3$  concentrations tend to be quite low, typically ranging from 20-40 ppb (NRC, 1991). It is reasonable to assume that, in the absence of anthropogenic emissions, the average summertime  $O_3$  concentration in the eastern half of the United States would be

## about 30-40 ppb (OTAG-AQA, 1997).

# $O_3$ Formation

While  $O_3$  formation in the atmosphere involves complex nonlinear processes, a simplified description of the process is offered here (Science and Technical Support Work Group, 1997).<sup>7</sup> Combustion sources that use air as an oxidizer will produce NO<sub>x</sub> when temperatures are above about 2200 degrees Celsius. In addition, incomplete combustion results in the emission of raw components and oxygenated organic components from the fuels. In sunlight these are sources of free radicals (e.g., OH, HO<sub>2</sub>, RO, RO<sub>2</sub>) that oxidize VOCs to carbonyls, CO, and carbon dioxide, while simultaneously oxidizing NO to NO<sub>2</sub> and recreating the free radical. Each free radical is cycled up to 5 times. The NO<sub>2</sub> reacts with sunlight to recreate NO and to produce O<sub>3</sub>. After the first oxidation of NO to NO<sub>2</sub>, every subsequent operation of the cycle produces an O<sub>3</sub> molecule with an efficiency of greater than 90 percent. In current chemical reaction mechanisms, a typical nitrogen is cycled 3 to 5 times. Some of the O<sub>3</sub> produced reacts with organics and with sunlight to produce more free radicals to maintain the cyclic oxidation process. This represents a powerful positive feedback process on the formation of more O<sub>3</sub>, given available NO<sub>x</sub>.

The carbonyls produced in the organic oxidation also react with sunlight to produce more free radicals. As the cycle operates,  $NO_2$  is converted into inorganic and organic nitrates; this form of nitrogen cannot cycle. This also removes free radicals. A system that converts all  $NO_x$  to nitrogen products cannot create any more  $O_3$ .  $NO_2$  reacts rapidly with free radicals and in situations that have a limited supply of radicals,  $NO_2$  can compete with the VOCs for the limited free radicals. This results in virtually no production of  $O_3$ . Large amounts of emitted NO relative to the radical sources prevents radical and NO cycling because the reaction between emitted NO and existing  $O_3$  removes  $O_3$  (a radical source), and the large amount of  $NO_2$  formed competes effectively with the VOCs for the other available radicals, thus leading to an overall suppression of  $O_3$  in such rich situations.

Different mixtures of VOCs and  $NO_x$ , therefore, can result in different  $O_3$  levels such that the total system is non-linear. That is, large amounts of VOCs and small amounts of  $NO_x$  make  $O_3$  rapidly but are quickly limited by removal of the  $NO_x$ . Decreases of VOCs under these circumstances show little effect on  $O_3$ . Large amounts of  $NO_x$  and small amounts of VOCs (which usually implies smaller radical source strengths) result in the formation of inorganic nitrates, but little  $O_3$ . In these cases, decreases in  $NO_x$  emissions result in an increase in  $O_3$ concentrations. Some combination of VOCs and  $NO_x$  is optimum at producing  $O_3$ .

The preceding is a static description. In the atmosphere, physical processes compete with chemical processes and change the outcomes in complex ways. The existence of feedback and non-linearity in the transformation system confound the description. Competing processes determine the ambient concentration and there are an infinite set of process magnitudes that can give rise to the same ambient concentrations and changes in concentrations. Lack of any direct

<sup>&</sup>lt;sup>7</sup> See, for example, NRC, 1991 for more information on O<sub>3</sub> chemistry.

measurement of process magnitudes result in the need to use inferential methods to confirm any one explanation of a particular  $O_3$  concentration.

#### Regional Scale of the $O_3$ Problem

As described in the preceding section, the impact of  $NO_x$  emissions on  $O_3$  concentrations is complex. While  $NO_x$  emissions are necessary for the formation of  $O_3$  in the lower atmosphere, a local decrease in  $NO_x$  emissions can, in some cases, increase local  $O_3$  concentrations. This effect of  $NO_x$  emissions reductions is further discussed in section IV. A of this document, Local and Regional  $NO_x$  Controls.

The formation of  $O_3$  is further complicated by biogenic emissions, meteorology, and transport of  $O_3$  and  $O_3$  precursors. The contribution of  $O_3$  precursor emissions from biogenic sources to local ambient  $O_3$  concentrations can be significant. This is especially true for emissions of biogenic VOCs. Important meteorological factors include temperature and wind direction and speed. Long-range transport of  $O_3$ , reactive N compounds, and partially oxidized organics such as aldehydes (which are excellent radical sources) can result in interactions between distant sources in urban or rural areas and local ambient  $O_3$ . Peroxyacetyl nitrate (PAN), formed from reaction of radicals with NO<sub>2</sub>, can transport NO<sub>x</sub> over relatively large distances through the atmosphere. Its rate of decomposition slows significantly with decreases in temperature, so that it can be formed near the surface in NO<sub>x</sub> rich areas, advected aloft to cooler conditions higher in the atmosphere, transported by the higher wind speeds aloft, and then be brought down to the warmer surface air to decompose and deliver NO<sub>2</sub> to downwind areas (NRC, 1991).

Typically,  $O_3$  episodes (periods including high  $O_3$  concentrations) last from 3-4 days on average, occur as many as 7-10 times a year, and often are of large spatial scale; in the eastern United States, high concentrations of  $O_3$  in urban, suburban, and rural areas tend to occur concurrently on scales of over 1000 kilometers (NRC, 1991). Maximum values of non-urban  $O_3$ commonly exceed 90 ppb during these episodes, compared with average daily maximum values of 60 ppb in summer. Thus, an urban area need contribute an increment of only 30 ppb over the regional background during a high  $O_3$  episode to cause a violation of the 1-hour  $O_3$  NAAQS of 120 ppb (NRC, 1991).

The precursors to  $O_3$  and  $O_3$  itself are transported long distances under some commonly occurring meteorological conditions. The transport of  $O_3$  and precursor pollutants over hundreds of kilometers can be a significant factor in the accumulation of  $O_3$  in any given area. Few urban areas in the United States can be treated as isolated cities unaffected by regional sources of  $O_3$  (NRC, 1991). As described below, there is a growing body of evidence that decreasing regional  $O_3$  levels by decreasing regional  $NO_x$  emissions holds the key to the ability of a number of the most seriously polluted nonattainment areas in the Eastern United States to attain and maintain the  $O_3$  NAAQS.

4. How much reduction in  $NO_x$  is needed nationally to achieve the  $O_3$  standard?

Summary: National, Regional, and Local Scale NO<sub>x</sub> Emissions Reductions Are Needed

As noted below, studies of the South, the Northeast Ozone Transport Region, and the states bordering Lake Michigan indicate that O<sub>3</sub> and O<sub>3</sub> precursors transported from attainment areas both within the regions and outside of the regions contribute to O<sub>3</sub> nonattainment within the regions. The extent of local controls that will be needed to attain and maintain the O<sub>3</sub> NAAQS in and near seriously polluted cities is sensitive both to the amount of  $O_3$  and precursors transported into the local area and to the specific photochemistry of the area. In some cases, preliminary local modeling with respect to the 1-hour standard performed by the states indicates that it may not be feasible to find sufficient local control measures for individual nonattainment areas unless transport into the areas is significantly decreased. The EPA has also conducted preliminary analyses for the new 8-hour O<sub>3</sub> standard which indicate that regional NO<sub>x</sub> emissions decreases would be effective in helping many areas attain that standard. These preliminary modeling studies consistently suggest that decreasing NO<sub>x</sub> emissions on a regional basis may be the most effective approach for decreasing  $O_3$  over large geographic areas, even though local NO<sub>x</sub> controls may be detrimental in urban centers of selected nonattainment areas on some days. Thus, large decreases in NO<sub>x</sub> emissions, in combination with other local controls, may be needed over much of the nation if all areas are to attain the  $O_3$  standard, as summarized below.

#### California

 $NO_x$  emissions reductions from 25-60 percent are needed in specific nonattainment areas. The State of California adopted their O<sub>3</sub> SIP on November 15, 1994. The SIP covers most of the populated portion of the state and relies on both NO<sub>x</sub> and VOC emissions reductions for most California nonattainment areas to demonstrate compliance with the NAAQS. Specifically, the revised SIP projects that the following NO<sub>x</sub> emissions reductions are needed (from a 1990 baseline): South Coast, 59 percent; Sacramento, 40 percent; Ventura, 51 percent; San Diego, 26 percent; and San Joaquin Valley, 49 percent.

The South Coast's control strategy for attainment of the  $O_3$  standard specifies a 59 percent decrease in  $NO_x$  emissions. The design of this strategy took into account the need to decrease  $NO_x$  as a precursor of PM, as described in the SIP submittal. This represents a decrease of over 800 tons of  $NO_x$  per day. The emissions reductions are to be achieved from a combination of national, state, and local control measures.

The Sacramento Metropolitan area's control strategy for attainment of the  $O_3$  standard specifies a 40 percent decrease in  $NO_x$  emissions. Modeling results indicate that  $NO_x$  emissions reductions in this urban area are more effective than VOC emissions reductions on a tonnage basis in lowering ambient  $O_3$  concentrations. The decreases are to be achieved from a combination of national, state, and local control measures, especially mobile source measures such as standards for heavy duty vehicles and off-road engines.

#### The Lake Michigan Area

Regional  $NO_x$  emissions reductions are needed. Modeling and monitoring studies performed to date for the states surrounding Lake Michigan (Wisconsin, Illinois, Indiana, and Michigan) indicate that decreasing  $O_3$  and  $O_3$  precursors transported into the nonattainment areas would have a significant effect on the number and stringency of local control measures to meet the 1-hour  $O_3$  NAAQS. In many cases boundary conditions appear to contribute significantly to peak  $O_3$  concentrations. The  $O_3$  and  $O_3$  precursors flowing into a metropolitan area can greatly influence the peak  $O_3$  concentration experienced in the metropolitan area. For example, the 1991 Lake Michigan Ozone Study found that transported  $O_3$  concentrations entering the region were 40-60 percent of the peak  $O_3$  concentrations in some of the metropolitan areas. The air mass entering the study area was measured by aircraft at 70-110 ppb on episode days (Roberts et al, 1994). In the Lake Michigan case, a 30 percent reduction in boundary conditions was shown by modeling to be as effective at lowering peak  $O_3$  concentrations as a 60 percent decrease in local VOC emissions (LADCO 1994).

These studies suggest that without such region-wide emissions reductions, the necessary degree of local control will be very difficult to achieve, even with very stringent local controls. The EPA Matrix study (Chu and Cox, 1995) reinforces that regional NO<sub>x</sub> control will be effective in lowering O<sub>3</sub> across the Midwest region. Taken together, the information available to date suggests that additional reductions in regional NO<sub>x</sub> emissions will probably be necessary in meeting the NAAQS in the Chicago/Gary/Milwaukee area and downwind (including western Michigan), even though currently available modeling shows that there may be a detrimental effect on some days from applying NO<sub>x</sub> controls locally in and near the major nonattainment areas.

#### New York Study

### Regional $NO_x$ emissions reductions of 75 percent are needed.

New York State's recent Urban Airshed Model (UAM) studies show that substantial decreases in the  $O_3$  transported from other regions would be necessary if several areas within the UAM domain are to achieve  $O_3$  attainment (John et al, 1994a,b). This UAM study demonstrates the potential effectiveness of a regional NO<sub>x</sub> emissions reduction strategy in combination with a local VOC emissions reduction strategy. The New York study showed that the combination of a regional strategy reflecting a 25 percent decrease in VOCs and a 75 percent decrease in NO<sub>x</sub> with a local strategy reflecting a 75 percent decrease in VOCs and a 25 percent decrease in NO<sub>x</sub> would be necessary for all areas throughout the New York UAM domain to lower predicted  $O_3$  levels to 120 ppb or less during adverse meteorological conditions.

#### Northeast Ozone Transport Region

#### Regional $NO_x$ emissions need to be decreased 50-75 percent.

In its analysis supporting the approval of a Low Emission Vehicle program in the mid-Atlantic and Northeast states comprising the Northeast Ozone Transport Region (OTR),<sup>8</sup> EPA reviewed existing work and performed analyses to evaluate in detail the degree to which  $NO_x$ controls are needed (EPA, 60 FR 48673). These studies indicated that  $NO_x$  emissions must be

<sup>&</sup>lt;sup>8</sup>The Northeast Ozone Transport Region (OTR) is comprised of the states of Maine, New Hampshire, Vermont, Massachusetts, Rhode Island, Connecticut, New York, New Jersey, Pennsylvania, Delaware, Maryland, and the Consolidated Metropolitan Statistical Area that includes the District of Columbia and northern Virginia.

decreased by 50 to 75 percent from 1990 levels to obtain predicted  $O_3$  levels of 120 ppb or less throughout the OTR. In addition, the EPA Matrix Study indicates that regional NO<sub>x</sub> emissions reductions are more effective at lowering high  $O_3$  concentrations than regional VOC emissions reductions.

More recent studies have confirmed these conclusions (Kuruville et al, 1994; Cox et al, 1993). Additional modeling simulations suggest that region-wide NOx controls coupled with urban-specific VOC controls would be needed for  $O_3$  attainment in the northeastern United States (Rao et al, 1995). Taken together, these studies point to the need to decrease  $NO_x$  emissions in the range of 50 to 75 percent throughout the OTR and that VOC emissions must also be decreased by the same amount in and near the Northeast urban corridor to reach and maintain predicted hourly maximum  $O_3$  levels of 120 ppb or less.

#### Southeast

 $NO_x$  emissions reductions up to 90 percent will be needed for the Atlanta area to attain. In the South, relatively high concentrations of  $O_3$  are measured in both rural and urban areas (Chameides and Cowling, 1995). Analyses of monitored data by Southern Oxidant Study investigators suggest that the background  $O_3$  levels are likely to be more responsive to decreases in NO<sub>x</sub> emissions than VOC emissions. Modeling to-date indicates that, in the absence of regional control measures, NO<sub>x</sub> emissions reductions on the order of ninety percent may be needed for the Atlanta area to attain the  $O_3$  standard during worst case weather conditions (Chameides and Cowling, 1995).

#### Ozone Transport Assessment Group (OTAG)

The EPA supported a consultative process among 37 eastern states which included examination of the extent to which  $NO_x$  emissions from as far as hundreds of kilometers away are contributing to smog problems in downwind cities in the eastern United States. Known as the OTAG and chaired by the State of Illinois, this group looked into ways of achieving additional cost-effective programs to further lower ground-level  $O_3$  throughout the eastern United States. The OTAG's modeling workgroup reported several key findings as a result of modeling analyses they conducted (OTAG-RUSM, 1997):

- \* NO<sub>x</sub> emissions reductions are more effective than VOC emissions reductions in lowering regional O<sub>3</sub> concentrations; NO<sub>x</sub> reductions decrease O<sub>3</sub> domainwide, while VOC reductions decrease O<sub>3</sub> only in urban areas.
- \* Elevated and low-level NO<sub>x</sub> emission reductions are both effective in lowering regional O<sub>3</sub> concentrations.
- \* More  $NO_x$  emission reductions result in more  $O_3$  benefit.
- \* Emission reductions in a given area mostly affect  $O_3$  in that same area.

- \* Emission reductions in a given area also affect  $O_3$  in downwind areas.
- \* Emission reductions will be effective in lowering 1-hour and 8-hour  $O_3$ .
- \* Regional reductions in  $NO_x$  emissions are necessary to help provide for attainment and maintenance of the  $O_3$  NAAQS in the eastern United States.

The OTAG's air quality analysis workgroup also reported several important findings as a result of analyses they conducted (OTAG-AQA, 1997):

- \* The distances of  $O_3$  impact deduced from multiple types of analysis range from 150 to 500 miles.
- \* The direct influence of specific urban areas can be directly traced to some 150-200 miles before merging indistinguishably into the regional  $O_3$  pattern.
- \*  $O_3$  transported at night can have a significant impact hundreds of miles downwind the next day.

# Integrated Strategies for Implementing the $O_3$ and PM Standards

#### **Common Factors**

As described in the sections on "Ozone" and "Particulate Matter," EPA published revisions to the  $O_3$  and PM NAAQS on July 18, 1997 (62 FR 38856). As part of the revisions process, EPA initiated action to address strategies for the implementation of the new NAAQS. These ongoing reviews and related implementation strategy activities to date have brought out important common factors between  $O_3$  and PM. Similarities in pollutant sources, formation, and control exist between  $O_3$  and PM, in particular the fine fraction of particles. These similarities provide opportunities for optimizing technical analysis tools (i.e., monitoring networks, emissions inventories, air quality models) and integrated emissions reduction strategies to yield important cross-cutting benefits across various air quality management programs. This integration could result in a net reduction of the regulatory burden on some source category sectors that would otherwise be impacted separately by  $O_3$ , PM, and visibility protection control strategies.

#### Federal Advisory Committee Act (FACA) Process

The EPA initiated a process designed to provide for significant stakeholder involvement in the development of integrated implementation strategies for the new/revised  $O_3$  and PM NAAQS and a new regional haze program. As described below, this process involves a new subcommittee of the Agency's Clean Air Act Advisory Committee (CAAAC), established in accordance with the FACA (5 U.S.C. App.2). The CAAAC was established to provide independent advice and counsel to the EPA on policy and technical issues associated with the implementation of the Act. The CAAAC advises EPA on the development, implementation, and enforcement of several of the new and expanded regulatory and market-based programs required by the Act.

The CAAAC advises on issues that cut across several program areas. A new subcommittee of the CAAAC, the Subcommittee for Ozone, Particulate Matter, and Regional Haze Implementation Programs (the Subcommittee), was established in August 1995 to address integrated strategies for the implementation of the new  $O_3$  and PM NAAQS, as well as a regional haze program. The focus of the Subcommittee will be on assisting EPA in developing implementation control strategies, preparing supporting analyses, and identifying and resolving impediments to the adoption of the resulting programs. The Subcommittee is composed of representatives selected from among state, local, and tribal organizations; environmental groups; industry; consultants; science/academia; and federal agencies. Recommendations made by the Subcommittee will be submitted to EPA through CAAAC.

5. How much reduction in  $NO_x$  emissions will be achieved with current and projected  $NO_x$  programs?

Substantial emissions reductions are currently being achieved through implementation of the CAA measures for mobile and stationary sources. These measures include the retrofit of reasonably available control technology on existing major stationary sources of  $NO_x$  and implementation of enhanced vehicle inspection and maintenance (I/M) programs under Title I, new tailpipe standards for new motor vehicles under Title II, and controls on certain coal-fired electric power plants under Title IV, Phase I. Total  $NO_x$  emissions will decline about 6 percent from current levels by the year 2000. Despite increases in vehicle miles traveled, total on-road vehicle emissions will likely continue to decline through 2005 as per vehicle emissions decrease due to tighter tailpipe standards, phase II reformulated gasoline is implemented, and I/M requirements are met.

Shortly after the year 2002, overall  $NO_x$  emissions are projected to begin to increase and continue to increase in the foreseeable future due to increased economic activity, unless new  $NO_x$  emissions reduction initiatives are implemented (EPA, 1995). It is clear that new controls will be needed to approach the decreases in  $NO_x$  emissions of 25-90 percent which are projected as being needed over large portions of the nation to attain the  $O_3$  standard. As described in Appendices A and B, several such new initiatives to decrease emissions of  $NO_x$  are planned or underway.

6. Summary

Emissions of NO<sub>x</sub> result in the formation of O<sub>3</sub> that can contribute to local O<sub>3</sub> nonattainment problems in some cases and/or, through long-range transport, contribute to nonattainment in downwind areas. High O<sub>3</sub> concentrations occur over large portions of the Eastern United States on some days during the summer. The transport of high O<sub>3</sub> concentrations into certain urban nonattainment areas makes it impractical for these urban areas to attain the NAAQS based on local controls alone. Thus, decreases in NO<sub>x</sub> emissions are needed locally in some areas to attain the O<sub>3</sub> NAAQS while, in other areas, NO<sub>x</sub> emissions reductions may be needed to help attain the O<sub>3</sub> NAAQS in downwind areas or to help maintain O<sub>3</sub> levels below the standard in attainment areas. Control strategies must consider efforts to decrease regional scale emissions as well as local emissions. In general,  $NO_x$  emissions reductions in upwind, rural areas coupled with VOC emissions reductions in urban nonattainment areas appears to be an effective strategy. In some cases however, the urban nonattainment area is also upwind of another urban nonattainment area or contains substantial biogenic VOC emissions. In these cases,  $NO_x$  emissions reductions may be needed in addition to, or instead of, VOC emissions reductions for purposes of  $O_3$  attainment. In both cases, decreases in precursor emissions in the upwind areas will help the downwind metropolitan areas attain and maintain the  $O_3$  standard. Thus, effective  $O_3$  control will require an integrated strategy that combines cost-effective emissions reductions in emissions at the local, state, regional, and national levels.

#### References

Chameides, W.L. and E.B. Cowling, *The State of the Southern Oxidant Study (SOS): Policy-Relevant Findings in Ozone Pollution Research, 1988-1994.* North Carolina State University, April 1995.

Chu, Shao-Hung and W.M. Cox, "Effects of Emissions Reductions on Ozone Predictions by the Regional Oxidant Model during the July 1988 Episode, *Journal of Applied Meteorology*, Vol. 34, Nol 3, March 1995.

Cox, William M. and Chu, Shao-Hung, "Meteorologically Adjusted Ozone Trends in Urban Areas: A Probabilistic Approach," *Atmospheric Environment*, Vol. 27B, No. 4, pp 425-434, 1993.

Finlayson-Pitts, B.J. and J.N. Pitts, Jr., "Atmospheric Chemistry of Tropospheric Ozone Formation: Scientific and Regulatory Implications," *Air and Waste Management Association*, Vol. 43, August 1993.

Georgia Environmental Protection Division, "The 1994 State Implementation Plan for the Atlanta Ozone Nonatttainment Area," 1994.

John, K., S.T. Rao, G.Sistla, W.Hao, and N.Zhou, 1994a, *Modeling Analyses of the Ozone Problem in the Northeast*, EPA-230-R-94-018, 1994.

John, K., S.T. Rao, G.Sistla, N.Zhou, W.Hao, K. Schere, S. Roselle, N.Possiel, R.Scheffe, 1994b, "Examination of the Efficacy of VOC and NOx Emissions Reductions on Ozone Improvement in the New York Metropolitan Area," printed in *Air Pollution Modeling and Its Application*, Plenum Press, NY, 1994.

Kuruville, John et al., *Modeling Analyses of Ozone Problem in the Northeast*, prepared for EPA, EPA Document No. EPA-230-R-94-108, 1994.

Lake Michigan Air Directors Consortium, Lake Michigan Ozone Study--Evaluation of the UAM-

# V Photochemical Grid Model in the Lake Michigan Region, 1994.

National Research Council, Committee on Tropospheric Ozone Formation and Measurement, *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, 1991.

Nichols, Mary D., Assistant Administrator for Air and Radiation, "Ozone Attainment Demonstrations," memorandum to EPA Regional Administrators, March 2, 1995.

Ozone Transport Assessment Group, "Summary of Modeling Data," prepared by Mike Koerber, Co-Chair Regional and Urban Scale Modeling Workgroup, for the June 2-3, 1997 Policy Group meeting.

Ozone Transport Assessment Group, "Summary of Air Quality Data," prepared by Dave Guinnup, Co-Chair Air Quality Analysis Workgroup, for the June 2-3, 1997 Policy Group meeting.

Rao, S.T., G. Sistla, W. Hao, K. John and J. Biswas, "On the Assessment of Ozone Control Policies for the Northeastern United States," presented at the 21st NATO/CCMS International Technical Meeting on Air Pollution Modeling and Its Application, Nov. 6-10, 1995.

Roberts, P.T., T.S. Dye, M.E. Korc, H.H. Main, *Air Quality Data Analysis for the 1991 Lake Michigan Ozone Study*, final report, STI-92022-1410-FR, Sonoma Technology, 1994.

Science and Technical Support Work Group, Ozone and Particulate Matter National Ambient Air Quality Standards, Federal Advisory Committee Act, Harvey Jeffries and Tom Helms, co-chairs, "Conceptual Model for Ozone, Particulate Matter and Regional Haze," February 19, 1997.

Seinfeld, John H., "Urban Air Pollution: State of the Science," February 10, 1989 vol., Science.

U.S. Environmental Protection Agency, Low Emission Vehicle Program for Northeast Ozone Transport Region; Final Rule, January 24, 1995 <u>Federal Register</u>, 60 FR 48673.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *National Air Pollutant Emission Trends*, 1900-1994," October 1995.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *Review of National Ambient Air Quality Standards for Ozone, Assessment of Scientific and Technical Information*, OAQPS Staff Paper, EPA-452/R-96-007, June 1996.

#### D. Particulate Matter

#### 1. Goals of the Program

The EPA has established health and welfare standards for particulate matter (PM). The goals of the program are to achieve and maintain these clean air standards throughout the country. As described below, emissions of  $NO_x$  can result in the formation of particulate nitrates that can contribute to PM nonattainment in some areas. Decreases in  $NO_x$  emissions might be needed in some areas to attain the PM NAAQS. In other areas,  $NO_x$  emissions reductions may not be needed to attain the PM NAAQS, but could help maintain PM levels below the standard in attainment areas.

#### 2. Status of the Programs

#### The NAAQS

Section 109 of the CAA directs the EPA Administrator to propose and promulgate primary and secondary NAAQS for pollutants identified under section 108. Section 109 defines a primary standard as that necessary to protect the public health, allowing an adequate margin of safety. A secondary standard, as defined in section 109, must specify an air quality concentration needed to protect the public welfare from any known or anticipated adverse effects associated with the presence of the pollutant in the ambient air. Welfare effects, as defined in section 302(h) of the CAA include, but are not limited to, effects on soils, water, crops, vegetation, materials, animals, wildlife, weather, visibility and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being.

States are primarily responsible for ensuring attainment and maintenance of the NAAQS. Under title I of the CAA, States are to submit, for EPA approval, SIPs that provide for the attainment and maintenance of such standards through control programs directed to sources of the pollutants involved. In addition, Federal programs provide for nationwide reductions in emissions of air pollutants through, for example, the New Source Performance Standards program under title I of the Act, which involves controls for major stationary sources.

#### РМ

The term PM refers to a solid or liquid material that is suspended in the atmosphere. PM includes materials of both organic and inorganic composition, and generally can also be divided into a primary component and secondary component. Primary PM consists of solid particles, aerosols, and fumes emitted directly as particles or droplets from various sources. Secondary PM is produced from gaseous pollutants, mainly SO<sub>2</sub>, NO<sub>x</sub>, ammonia, and some VOCs. These

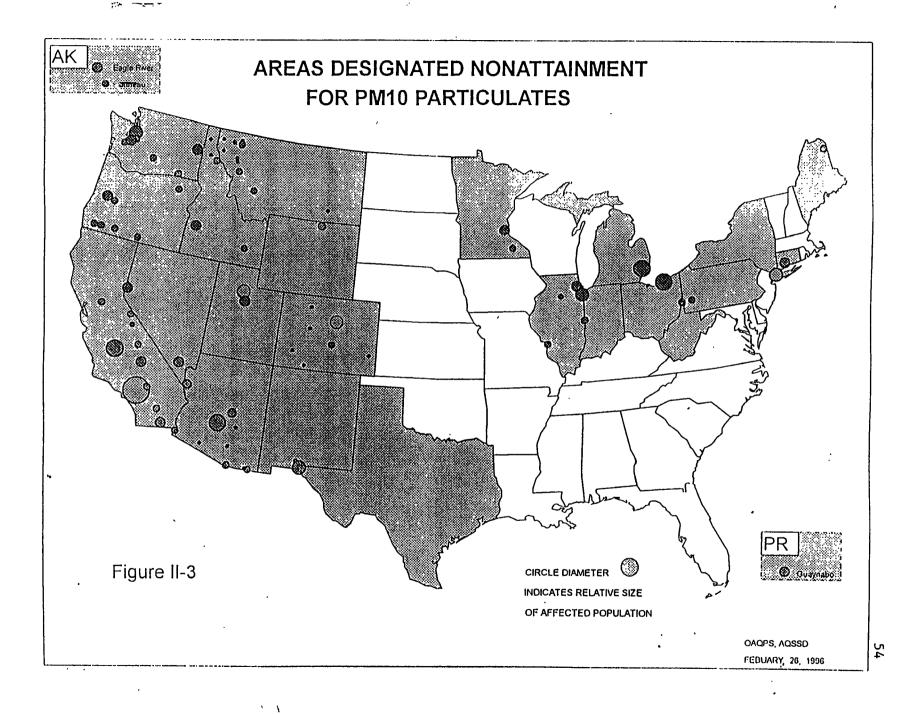
precursor gases react with one another and with oxygen and water in the atmosphere to form particles or condensible compounds. The chemical and physical properties of PM vary greatly with time, region, meteorology, and source category, thus complicating their understanding and control.

#### The PM NAAQS

The PM NAAQS include  $PM_{2.5}$  standards and  $PM_{10}$  standards. The  $PM_{2.5}$  standards are set at 15 micrograms per cubic meter, annual mean, and 65 micrograms per cubic meter, 24-hour average. The  $PM_{10}$  standards are set at 50 micrograms per cubic meter, annual average, and 150 micrograms per cubic meter, 24-hour average. (For more details see the "Most Recent Review of the Particulate Matter NAAQS" section below).

#### Areas That Do Not Meet the PM<sub>10</sub> NAAQS

In 1990 EPA designated 70 areas as moderate nonattainment for  $PM_{10}$ , and 13 additional areas were added in 1994 for a total of 83  $PM_{10}$  nonattainment areas. Five of the initial areas have been reclassified to serious nonattainment areas. Based on air quality data for 1992 to 1994, 37 of these (but none of the serious areas) were determined to have met the  $PM_{10}$  NAAQS by their December 31, 1994 attainment date. The current 46 nonattainment areas are shown in Figure II-3 below.



# Establishment of the PM NAAQS and Subsequent Reviews Establishment of the NAAQS for PM

NAAQS for PM were first established in 1971 (April 30, 1971<u>Federal Register</u>). The reference method specified for determining attainment of the original standards was the high-volume sampler, which collects PM up to a nominal size of 25 to 45 microns (so-called total suspended particulate or TSP). The primary standards (measured by the indicator TSP) were 260 micrograms per cubic meter, 24-hour average, not to be exceeded more than once per year, and 75 micrograms per cubic meter, annual geometric mean. The secondary standard (measured as TSP) was 150 micrograms per cubic meter, 24-hour average not to be exceeded more than once per year.

#### First Review of NAAQS for PM

In October 1979 (44 FR 56731), EPA announced the first review of the criteria document and NAAQS for PM and, after a lengthy and elaborate process, promulgated significant revisions of the original standards in 1987 (52 FR 24854, July 1, 1987). In that decision, EPA changed the indicator for particles from TSP to  $PM_{10}$ , the latter referring to particles with a mean aerodynamic diameter less than or equal to 10 microns.<sup>9</sup> EPA also revised the acceptable concentration and form of the primary standards by 1) replacing the 24-hour TSP standard with a 24-hour  $PM_{10}$  standard of 150 micrograms per cubic meter with no more than one expected exceedance per year averaged over 3 years and 2) replacing the annual TSP standard with a  $PM_{10}$  standard of 50 micrograms per cubic meter, expected annual arithmetic mean. The secondary standard was revised by replacing it with 24-hour and annual standards identical in all respects to the primary standards. The revisions also included a new reference method for the measurement of  $PM_{10}$  in the ambient air and rules for determining attainment of the new standards.

# Most Recent Review of the PM NAAQS

To initiate its most recent review, EPA analyzed thousands of peer-reviewed scientific studies. These studies were then synthesized, along with a recommendation on whether the existing standards were adequately protective, and presented to an independent scientific advisory body ("CASAC"), as required by the CAA. After holding more than 125 hours of public discussion, and based upon 250 of the most relevant studies, CASAC concluded that EPA's current  $O_3$  and particulate standards should be strengthened. This review took several years to complete.

On December 13, 1996, EPA proposed in the <u>Federal Register</u> to change the PM standard (61 FR 65638). As described in detail in that notice, EPA proposed to change the current standards by adding two new primary  $PM_{2.5}$  standards set at 15 micrograms per cubic

<sup>&</sup>lt;sup>9</sup>The more precise term is 50 percent cut point or 50 percent diameter. This is the aerodynamic particle diameter for which the efficiency of particle collection is 50 percent. Larger particles are not excluded altogether, but are collected with substantially decreasing efficiency and smaller particles are collected with increasing (up to 100 percent) efficiency. Ambient samplers with this cut point provide a reliable estimate of the total mass of suspended particulate matter of aerodynamic size less than or equal to 10 microns.

meter, annual mean, and 50 micrograms per cubic meter, 24-hour average. The revisions would provide increased protection against a wide range of potential PM-related health effects. The proposed annual  $PM_{2.5}$  standard would be based on the 3-year average of the annual arithmetic mean  $PM_{2.5}$  concentrations, spatially averaged across an area. The proposed 24-hour  $PM_{2.5}$  standard would be based on the 3-year average of the 98th percentile of 24-hour  $PM_{2.5}$  concentrations at each monitor within an area. The EPA proposed to revise the current 24-hour  $PM_{10}$  standard of 150 micrograms per cubic meter by replacing the 1-expected-exceedance form with a 98th percentile form, averaged over 3 years at each monitor within an area. The EPA proposed to revise the current secondary standards by making them identical to the suite of proposed primary standards.

EPA then conducted an extensive public comment process, receiving approximately 57,000 comments at public hearings held across the country and through written, telephone and computer messages. The proposed standards were also subjected to an intensive inter-agency review process. A court order required EPA to finalize a PM standard by mid-July of this year, and EPA committed to a court to do the same for  $O_3$ .

EPA's final air quality standards for  $O_3$  and PM were published in the <u>Federal Register</u> of July 18, 1997 (62 FR 38856). With respect to PM, the final standards include one significant change from EPA's 1996 proposal: the final standard set the 24-hour limit at 65 micrograms per cubic meter, instead of 50 micrograms (as proposed), to provide maximum flexibility for local areas and sources, while still retaining the public health protections of the proposal that are incorporated into the annual standard.

3. Science of NO<sub>x</sub> and PM

#### Health and Welfare Effects

Exposure to airborne PM has a wide range of adverse health effects. The damages caused by PM vary depending on its concentration, composition, and the sizes of the constituent particles. A summary of these effects is provided below; for further information, see EPA's notice of proposed rulemaking on "National Ambient Air Quality Standards for Ozone and Particulate Matter" published in the December 13, 1997 <u>Federal Register</u> and relevant documents referenced in that notice.

As discussed in EPA's Criteria Document (EPA, April 1996) and Staff Paper (EPA, July 1996) and summarized in the December 13, 1996 proposal notice, the key health effects associated with PM include: 1) premature mortality; 2) aggravation of respiratory and cardiovascular disease (as indicated by increased hospital admissions and emergency room visits, school absences, work loss days, and restricted activity days); 3) changes in lung function and increased respiratory symptoms; 4) changes to lung tissues and structure; and 5) altered respiratory defense mechanisms. Most of these effects have been consistently associated with ambient PM concentrations, which have been used as a measure of population exposure, in a number of community epidemiological studies. Although mechanisms by which particles cause

effects have not been elucidated, there is general agreement that the cardio-respiratory system is the major target of PM effects.

The EPA revised the secondary (welfare-based) PM NAAQS by making them identical to the primary standards. The EPA believes that the  $PM_{2.5}$  and  $PM_{10}$  standards, combined with the CAA required regional haze program, will provide protection against the major PM-related welfare effects. These welfare effects include visibility impairment, soiling, and materials damage. The Administrator of EPA signed the proposed rulemaking notice for the regional haze rules on July 18, 1997.

# Size of Particles

The health and environmental effects of PM are strongly related to the size of the particles (EPA Staff Paper, 1996). The aerodynamic size and associated composition of particles determines their behavior in the respiratory system (i.e., how far the particles are able to penetrate, where particles are deposited, and how effective the body's clearance mechanisms are in removing them). Furthermore, particle size is one of the most important parameters in determining atmospheric lifetime of particles, which is a key consideration in assessing health effects information because of its relationship to exposure. The total surface area and number of particles, chemical composition, water solubility, formation process, and emission sources all vary with particle size. Particle size is also a determinant of visibility impairment, a welfare consideration linked to fine particle concentrations. Thus, size is an important parameter in characterizing PM, and particle diameter has been used to define the present standards.

#### Atmospheric Behavior of Fine and Coarse Particles

Sulfates, nitrates, and some organic particles as well as their precursors can remain in the atmosphere for several days and can be carried hundreds or even thousands of kilometers from their sources to remote locations such as national parks and wilderness areas (NRC, 1993). Fine particles are small enough that gravitational forces are largely overcome by the random forces from collisions with gas molecules. Thus fine particles tend to follow air streams and are difficult to remove by impaction on surfaces. Therefore, fine particles have very long lifetimes in the atmosphere, travel long distances, and tend to be more uniformly distributed over larger geographic areas than coarse particles (EPA, 1996). The atmospheric lifetimes of fine particles with respect to dry deposition is on the order of weeks. Removal of fine particles occurs when the particles absorb water, grow into cloud droplets, grow further to rain drops, and fall out as rain. This process lowers the atmospheric lifetime of fine particles to on the order of several days.

In contrast, coarse particles are large enough so that the force of gravity exceeds the buoyancy forces of the surrounding air currents leading to their settling out to the earth's surface. Coarse particles are in the 2.5 to 10 micron size range. These larger particles tend to fall rapidly out of the air, with atmospheric lifetimes of only minutes to hours depending on their size. Coarse particles are also too large to follow air streams, such that they tend to be easily removed by

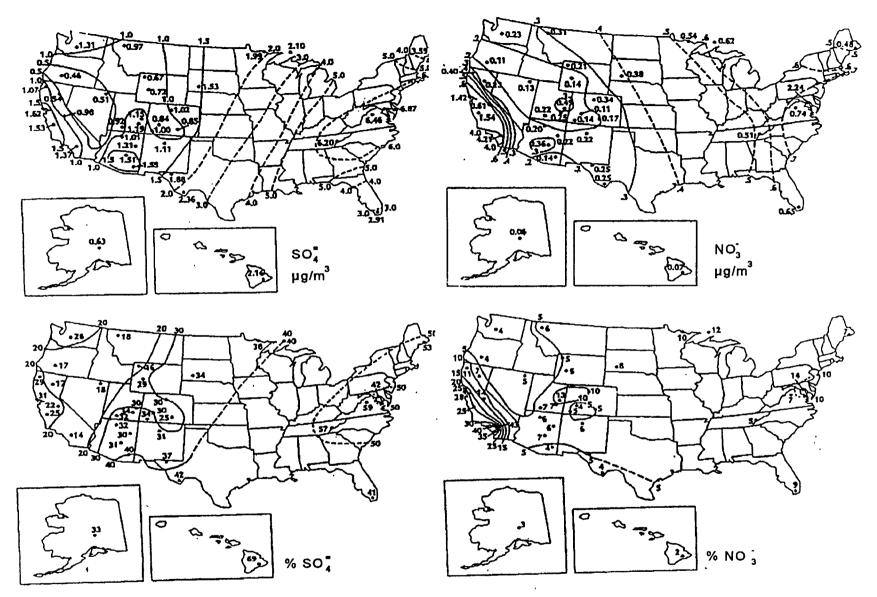
impaction on surfaces. Coarse particles are primarily composed of crustal elements (silicon, aluminum, iron and potassium); biological materials (bacteria, pollen, and spores) also appear in the coarse mode.

#### **Emission Sources and Formation Processes of Particles**

In most locations, a variety of diverse activities contribute significantly to PM concentrations, including fuel combustion (from vehicles, power generation, and industrial facilities), residential fireplaces, agricultural and silvicultural burning, and atmospheric formation from gaseous precursors (largely produced from fuel combustion). Other sources include construction and demolition activities, wind blown dust, and road dust. From these diverse sources come the mix of substances that comprise PM. The major chemical constituents of  $PM_{10}$  are sulfates, nitrates, carbonaceous compounds (both elemental and organic carbon compounds), acids, ammonium ions, metal compounds, water, and crustal materials. The amounts of these components vary from place to place and over time.

Coarse particles are primarily the result of crushing or grinding processes. Fine particles result from (1) direct emissions, (2) gaseous emissions which condense in the atmosphere without any other chemical reactions, and (3) precursor gases that later chemically react to form fine particles. Particles formed as a result of chemical reaction of gases are termed secondary particles because the direct emissions from a source is a gas (e.g.,  $SO_2$  or NO) that is subsequently converted to a low vapor pressure substance in the atmosphere. Sources of fine and coarse particles are summarized in Tables II-2 and II-3 (EPA, 1996). The fraction of fine particulate due to sulfate is greater in the East, and the nitrate fraction is larger in the West (see figure II-4; EPA, April 1996).

Transformation from gases to particles requires substantial interaction in the atmosphere. Such transformation can take place locally, during prolonged stagnations, or during transport over long distances. Moisture, sunlight, temperature, and the presence or absence of fogs and clouds affect transformation. In general, particles formed from these types of secondary processes will be more uniform in space and time than those that result from primary emissions.



Yearly average absolute and relative concentrations for sulfate and nitrate.

١

Source: Sisler et al. (1993) and Malm et al. (1994b).

.

.

A large fraction of the mass in the fine size fraction is derived from material that has been volatilized in combustion chambers and then recondensed to form primary fine PM, or has been formed in the atmosphere from precursor gases as secondary PM. Since precursor gases and fine PM are capable of traveling great distances, it is difficult to identify precisely the contribution of the individual sources. Sulfuric acid, which is the source of particle strong acidity and sulfates, is formed from the atmospheric reaction of SO<sub>2</sub> which is formed during combustion of sulfur compounds contained in fossil fuels. As noted below, nitrates are formed by atmospheric reactions of NO<sub>x</sub> which are generated during combustion or other high temperature processes. Ammonia, which neutralizes sulfuric and nitric acid to form sulfates and nitrates, has a variety of sources, the most important being emissions from animal waste and fertilizers.

PM may be formed from emissions of NO which are converted to NO<sub>2</sub> which then participates in various reactions to form other substances, including O<sub>3</sub> and PM. Nitrate airborne particles can be produced by several mechanisms. One major mechanism of nitrate formation involves nitric acid vapor which has a much higher vapor pressure than sulfuric acid and tends to stay more in the gas phase. Nitric acid (HNO<sub>3</sub>) is mostly formed in the gas-phase reaction of NO<sub>2</sub> with the hydroxyl radical. The gaseous nitric acid can react with ammonia to form ammonium nitrate or at airborne particle surfaces to form nitrate salts, such as sodium nitrate. Thus, nitrate size distributions depend, in part, on the size distributions of the particles on which they react. Conditions that favor aerosol nitrate formation include high nitric acid concentrations, high ammonia (gas phase) or salt particle concentrations, low temperatures, and high relative humidity. If the air parcel carrying the aerosol nitrate experiences a temperature increase and/or decrease in humidity, the concentration of the aerosol nitrate would be expected to decline as the nitric acid or ammonia returns to gas phase. Fine particle nitrate concentrations near 100 micrograms per cubic meter over 24-hour averaging times have been observed in the eastern end of the South Coast Air Basin that surrounds Los Angeles during late October (Science and Technical Support Work Group, 1997).

## Visibility-Impairing Particles

As described in the "Visibility Protection" section of this document, fine particles are effective in impairing visibility by scattering or absorbing light. Different types of particles have varying efficiencies in causing visibility impairment. The fine particles principally responsible for visibility impairment are sulfates, nitrates, organic matter, elemental carbon (soot), and soil dust. Coarse particles also impair visibility, although less efficiently than fine particles.

		Source	25			
Primary PM			Second	Secondary PM		
Aerosol species	Natural	Anthropogenic	Natural	Anthropogenic		
SO4 <sup>=</sup>	Sea spray	Fossil fuel combustion	Oxidation of reduced sulfur gases emitted by the oceans and wetlands; and $SO_2$ and $H_2S$ emitted by volcanism and forest fires	Oxidation of SO <sub>2</sub> emitted from fossil fuel combustion		
NO <sub>3</sub> -	_	Motor vehicle exhaust	Oxidation of NO <sub>x</sub> produced by soils, forest fires, and lighting	Oxidation of NO <sub>x</sub> emitted from fossil fuel combustion; and in motor vehicle exhaust		
Minerals	Erosion, re-entrainment	Fugitive dust; paved, unpaved roads; agriculture and forestry	_	_		
$\mathrm{NH_4^+}$	_	Motor vehicle exhaust	Emissions of NH <sub>3</sub> from wild animals, undisturbed soil	Emissions of NH <sub>3</sub> from animal husbandry, sewage, fertilized land		
Organic carbon	Wild fires	Open burning, wood burning, cooking, motor vehicle exhaust, tire wear	Oxidation of hydrocarbons emitted by vegetation, (terpenes, waxes); wild fires	Oxidation of hydrocarbons emitted by motor vehicles, open burning, wood burning		
Elemental carbon	Wild fires	Motor vehicle exhaust, wood burning, cooking	_	_		
Metals	Volcanic activity	Fossil fuel combustion, smelting, brake wear	_	_		
Bioaerosols	Viruses, bacteria	_	_	_		

TABLE II-2 CONSTITUENTS OF ATMOSPHERIC FINE PARTICLES LESS THAN 2.5 MICRONS AND THEIR MAJOR SOURCES

Sources							
	Primary	Secondary					
Aerosol species	Natural	Anthropogenic	Natural	Anthropogenic			
Minerals	Erosion, re-entrainment	Fugitive dust; paved, unpaved road dust, agriculture and forestry	_	_			
Metals	Erosion, re-entrainment, organic debris	_	_	_			
Miscellaneous ions	Sea spray	Road salting	—	_			
Organic carbon	_	Tire and asphalt wear	_	_			
Organic debris	Plant, insect fragments	_	—	_			
Bioaerosols	Pollen, fungal spores, bacterial agglomerates		_	_			

# TABLE II-3 CONSTITUENTS OF ATMOSPHERIC PARTICLES GREATER THAN 2.5 MICRONS AND THEIR MAJOR SOURCES

#### 4. How much reduction is needed?

#### Implementing the PM<sub>10</sub> Standards

As shown in figure II-3, there are still several  $PM_{10}$  nonattainment areas in the country. Some of these areas may need to consider decreases of  $NO_x$  emissions as part of their attainment planning. The importance of  $NO_x$  as a  $PM_{10}$  precursor varies significantly from place-to-place.

#### Integrated Strategies for Implementing the O<sub>3</sub> and PM Standards Common Factors

As noted above, EPA published revisions to the  $O_3$  and PM NAAQS on July 18, 1997. As part of the revisions process, EPA initiated action to address strategies for the implementation of the new NAAQS. These ongoing reviews and related implementation strategy activities to date have brought out important common factors between  $O_3$  and PM. Similarities in pollutant sources, formation, and control exist between  $O_3$  and PM, in particular the fine fraction of particles. These similarities provide opportunities for optimizing technical analysis tools (i.e., monitoring networks, emissions inventories, air quality models) and integrated emissions reduction strategies to yield important cross-cutting benefits across various air quality management programs. This integration could result in a net reduction of the regulatory burden on some source category sectors that would otherwise be impacted separately by  $O_3$ , PM, and visibility protection control strategies.

#### Federal Advisory Committee Act (FACA) Process

The EPA initiated a process designed to provide for significant stakeholder involvement in the development of integrated implementation strategies for the new/revised  $O_3$  and PM NAAQS and a new regional haze program. As described below, this process involves a new subcommittee of the Agency's Clean Air Act Advisory Committee (CAAAC), established in accordance with the FACA (5 U.S.C. App.2). The CAAAC was established to provide independent advice and counsel to the EPA on policy and technical issues associated with the implementation of the Act. The CAAAC advises EPA on the development, implementation, and enforcement of several of the new and expanded regulatory and market-based programs required by the Act.

The CAAAC advises on issues that cut across several program areas. A new subcommittee of the CAAAC, the Subcommittee for Ozone, Particulate Matter, and Regional Haze Implementation Programs (the Subcommittee), was established in August 1995 to address integrated strategies for the implementation of the new  $O_3$  and PM NAAQS, as well as a regional haze program. The focus of the Subcommittee will be on assisting EPA in developing implementation control strategies, preparing supporting analyses, and identifying and resolving impediments to the adoption of the resulting programs. The Subcommittee is composed of representatives selected from among state, local, and tribal organizations; environmental groups; industry; consultants; science/academia; and federal agencies. Recommendations made by the Subcommittee will be submitted to EPA through CAAAC.

5. How much reduction will be achieved with current and projected programs?

As described in detail in the Appendices, several current and future programs will achieve decreases in  $NO_x$  emissions. In addition, some States are planning or have underway  $PM_{10}$  attainment plans which specifically call for  $NO_x$  emissions reductions in certain areas, including, for example, the South Coast Air Basin in California. Additional  $NO_x$  emissions reductions might be needed to attain the new  $PM_{2.5}$  standards.

6. Summary

Emissions of NO<sub>x</sub> result in the formation of particulate nitrates which contribute to  $PM_{10}$  nonattainment in some areas. Decreases in NO<sub>x</sub> emissions are needed in some areas to attain the  $PM_{10}$  NAAQS, including the Los Angeles area. In other areas, NO<sub>x</sub> emissions reductions may be needed to attain the  $PM_{2.5}$  NAAQS and/or help maintain concentrations below the PM NAAQS in attainment areas.

# References

National Research Council, Committee on Haze in National Parks and Wilderness Areas *Protecting Visibility in National Parks and Wilderness Areas*. National Academy Press, Washington, D.C., 1993.

Science and Technical Support Work Group, Ozone and Particulate Matter National Ambient Air Quality Standards, Federal Advisory Committee Act, Harvey Jeffries and Tom Helms, co-chairs, "Conceptual Model for Ozone, Particulate Matter and Regional Haze," February 19, 1997.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards Air Quality *Criteria for Particulate Matter* (Criteria Document) (three volumes, EPA/600/P-95-001aF thru EPA/600/P-95-001cF, April 1996, NTIS # PB-96-168224.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *Review of the national ambient air quality standards for particulate matter: policy assessment of scientific and technical information*, (Staff Paper), EPA-452/R-96-013, July 1996, NTIS # PB-97-115406.

#### E. Visibility Protection

#### 1. Goals of the Program

Visibility is an air quality related value essential to the enjoyment of national parks, wilderness areas, and other scenic areas throughout our country. In section 169A of the 1977 amendments to the CAA, Congress recognized that visibility was an important aspect of public welfare that should be protected. It established as a national goal "the prevention of any future, and the remedying of any existing, impairment of visibility in mandatory class I Federal areas which impairment results from man-made air pollution." The CAA also calls for the development of programs to ensure reasonable progress toward the national goal, including the establishment of a new regional haze program for the protection of visibility in mandatory Federal Class I areas across the country. These programs are to be implemented by the States and can be regionally specific.

Regional haze and other visibility impairment is primarily caused by fine particles in the air which scatter or absorb light. These particles include, elemental carbon (soot), nitrates, organic matter, soil dust, and sulfates. The major cause of visibility impairment in the Eastern United States is sulfate, formed primarily from  $SO_2$  emitted from coal combustion by electric utility boilers, while in the West the other four particle types play a greater role. Emissions of  $NO_x$  lead to the formation of particulate nitrates. Thus, the decreases in emissions of  $NO_x$  will help improve visibility and make progress toward the national goal.

2. Status of the Program

#### CAA Visibility Requirements

The CAA includes two emissions control programs specifically concerned with visibility in national parks and wilderness areas. One of these, the Prevention of Significant Deterioration program is directed mainly at new sources, as noted below. The other, a visibility protection program, is aimed largely at existing sources.

#### *Prevention of Significant Deterioration (PSD)*

The PSD program requires each new or expanded major emitting facility locating in a clean air area to install the best available control technology and meet increments that limit the cumulative increases in pollution in clean air areas. Because all areas of the country are clean with respect to the  $NO_2$  standard, the PSD program applies to major sources of  $NO_x$  throughout the nation.

The PSD program has protected visibility to some extent by decreasing the growth of emissions of pollutants that contribute to regional haze, including  $SO_2$  and  $NO_x$ . The PSD

program includes a special air quality related values (AQRV) test for evaluating a major emitting facility that might affect a Class I area. Many large national parks and wilderness areas are designated as Class I areas and therefore are subject to the most stringent increments. The federal land manager has the responsibility to protect the AQRV. The EPA's PSD regulations are found in the <u>Code of Federal Regulations</u> at 40 CFR 52.21.

#### Visibility Rules

The EPA issued visibility rules in 1980 (See 40 CFR 51.300-307 and 45 FR 80084) requiring states containing mandatory class I areas to: (1) develop a program to assess and remedy visibility impairment from new and existing sources; (2) develop a long-term strategy to assure progress toward the national goal; (3) develop a visibility monitoring strategy; (4) consider "integral vistas" outside of class I areas in all aspects of visibility protection; and (5) notify Federal land managers (FLM) of proposed new major stationary sources and consider visibility analyses conducted by FLMs in their permitting decisions (40 CFR 51 subpart P). These visibility rules lay out a process for visibility impacts from a single source or small group of sources which may reasonably be anticipated to cause or contribute to visibility impairment in a Federal Class I area. The process is initiated by a certification of impairment by a FLM. The State determines whether the impairment is attributable to the source(s) and, if so, requires controls to reduce impairment.

The 1980 visibility regulations required 36 States containing mandatory Class I areas to submit strategies for monitoring visibility. The EPA completed in 1985 Federal regulations to establish a national visibility monitoring network which would be cooperatively managed by EPA, Federal land management agencies, and State air agency representatives. This network is now known as IMPROVE (Interagency Monitoring of Protected Visual Environments). Due to resource limitations, IMPROVE monitors could not be placed in all 156 mandatory class I areas. Instead, the IMPROVE Steering Committee has selected a set of priority sites. Data are currently being collected at more than 40 Class I locations. The IMPROVE monitoring protocol specifies aerosol, photographic, and optical (light extinction) measurements twice a week.

At that time of the rulemaking, EPA also expressed its intention to regulate regional haze at some future date when monitoring techniques are improved and the relationship between air pollutants and visibility impairment is better established. Much progress has been made in technical areas important to the successful implementation of a regional haze program, including areas such as visibility monitoring, regional scale modeling, and scientific knowledge of the regional effects of particles on visibility. As described below, EPA plans to begin a new regional haze program for the protection of visibility in mandatory Federal Class I areas<sup>10</sup> across the country.

#### Grand Canyon Visibility Transport Commission

Section 169B of the 1990 CAA amendments required the establishment of the Grand

<sup>&</sup>lt;sup>10</sup>Areas designated as mandatory class I areas are those national parks exceeding 6000 acres, wilderness areas and memorial parks exceeding 5000 areas, and all international parks which were in existence on August 7, 1977.

Canyon Visibility Transport Commission (GCVTC<sup>11</sup>). The Commission was charged with assessing adverse impacts on visibility from projected growth in the region, and requires the Commission to recommend measures to EPA for addressing adverse impacts to visibility in the region. The Commission formally adopted its report to the EPA on June 10, 1996. The EPA is to use the Commission's recommendations as guidance for developing national strategies and/or rulemaking. Implementation of specific program components will be the responsibility of tribes, states, and, in some cases, federal agencies. The primary recommendations in the Commission's report include: air pollution prevention measures such as energy conservation and increased energy efficiency; tracking of emissions growth in clean air corridors; development of a plan for an emissions cap and trading program for stationary sources; and establishing a regional emissions budget for mobile sources.

#### **Regional Haze Rules**

The Administrator of EPA signed the notice of proposed rulemaking for the regional haze rules on July 18, 1997. These rules are a continuation of the 1980 rules. From the time the GCVTC report is received, section 169B requires EPA to issue rules within 18 months to assure reasonable progress toward remedying adverse impacts due to regional haze. After establishment of a regional haze program, States affected by these rules are required, under section 169B(e)(1), to revise their state implementation plans (SIPs) within 12 months to include such emission limits, schedules of compliance, and other measures as may be necessary for program implementation.

#### Relation to NO<sub>2</sub> NAAQS Review

As described in the final rulemaking notice of October 8, 1996 (61 FR 52852) on revision to the NO<sub>2</sub> NAAQS, EPA determined that establishment of a secondary NO<sub>2</sub> standard to protect visibility is not appropriate. While NO<sub>2</sub> can contribute to brown haze, there is no established relationship between ground level NO<sub>2</sub> concentrations at a given point and visibility impairment due to a plume or regional haze. Furthermore, regional scale NO<sub>2</sub> light extinction is much less than aerosol extinction. These considerations helped lead to the conclusion that establishment of a secondary NO<sub>2</sub> standard to protect visibility would not be appropriate.

#### Relation to PM Standard Review

In reviewing the NAAQS for PM, EPA also considered the appropriateness of a secondary standard to address a number of welfare effects, specifically including visibility. Because of regional variations in visibility conditions created by background concentrations of fine particles, annual average humidity, pollutant mix, and resulting total light extinction, however, a regional haze program under the regulatory authority in section 169A may be preferable to setting a secondary NAAQS. As described in the July 18, 1997 <u>Federal Register</u>,

<sup>&</sup>lt;sup>11</sup>The Commission consists of the Governors, or their designees, from the States of Arizona, California, Colorado, Nevada, New Mexico, Oregon, Utah, and Wyoming; the President of the Navajo Nation; the Chairman of the Hopi Tribe; the Governor of the Pueblo of Acoma; the Chairman of the Hualapai Tribe; and ex-officio members from EPA, Bureau of Land Management, National Park Service, United States Forest Service, United States Fish and Wildlife Service, and the Columbia River Inter-Tribal Fish Commission.

EPA set the secondary PM standards identical to the primary standards which, in conjunction with a regional haze program under sections 169A and 169B of the Act, EPA believes is the most appropriate and effective means of addressing the welfare effects associated with visibility impairment. Together, the two programs and associated control strategies should provide appropriate protection against the effects of PM on visibility and allow all regions of the country to make reasonable progress toward the national visibility goal.

3. Science of NO<sub>x</sub> and Visibility

#### Visibility Basics

For an object to be seen against a background, there must be sufficient contrast between the object and its background. That is, the light from the object and the background must be sufficiently different in apparent brightness or color to make the object stand out against the background. Light from objects and their background viewed through the atmosphere from a distance are modified by the particles and gases in the atmosphere (primarily clean air).

Light as it traverses the atmosphere is scattered (i.e., redirected in directions) and absorbed (i.e., converted from light to heat) by the particles and gases in the atmosphere. This affects the appearance of scenes in two ways. The image-forming light from scenic features is diminished since a fraction of the light is scattered or absorbed; non-image-forming light is scattered into the sight path. Both of these effects lower the contrast between object and background, and cause the scene to be more obscured. This decrease in contrast is further decreased with distance to the scenic feature being viewed, and with increased concentration of airborne particles.

Many studies have been published on visibility conditions and related aerosol concentrations. The NAPAP report (1991) lists 33 aerosol and visibility databases. From these studies, the major contributors to visibility impairment from natural and man-made sources are sulfate particles, organic particles, elemental carbon, suspended dust, and nitrate particles.

Regional haze is primarily caused by fine particles in the air, typically less than 2.5 microns in diameter, which scatter and absorb light. These particles include sulfates, nitrates, organic matter, elemental carbon (soot), and soil dust (NRC, 1993). The fate of regional haze is a function of meteorological and chemical processes, sometimes causing fine particle loadings to remain suspended in the atmosphere for long periods of time (3-5 days) and to be transported long distances (thousands of kilometers) from their sources. A large fraction of anthropogenic airborne particles (sulfates, nitrates, and some organic particles) accumulates in the 0.1-1.0 micron diameter range. These particles can survive in the atmosphere for several days and can be transported hundreds or even thousands of kilometers from their sources to remote locations, such as national parks and wilderness areas (NRC, 1993). During transport, the emissions from many different sources can become mixed, making it difficult to assess the effects of individual sources on visibility.

# Types of Visibility Impairment

Visibility effects are manifested in two principal ways: (1) as local impairment (e.g., plumes and localized hazes) and (2) as regional haze. Local-scale impairment is defined as impairment that is "reasonably attributable" to a single source or group of sources. Visibility impairment in some urban areas can be dominated by local rather than regional sources, particularly in mountain valleys in the winter and in meteorologically stagnant conditions.

The second category of impairment, regional haze, is produced from a multitude of sources combined over many days. Regional haze impairs visibility in every direction over a large area. Objects on the horizon are obscured and the texture of nearby objects is reduced. In some cases, the haze may be elevated and appear as layers of discoloration.

The contribution by particles from both natural and man-made sources are highly variable. Natural particle sources such as wildfires, windblown dust, salts from ocean spray, etc. are highly variable across time and space with the result that natural background levels of visibility are highly variable. Concentrations of man-made and natural particles also vary because of the influence of variable meteorology responsible for atmospheric transport and dispersion. (For further information on visibility, see the 1979 Report and the 1990 NAPAP Report "Acidic Deposition: State of Science and Technology, Volume III, Report Number 24.)

#### Visibility Metrics

Visual range, which is defined as the greatest distance that a large dark object can be seen against the background sky, is the oldest and most commonly used visibility metric. Visual range was developed and continues to function well as an aid in military operations and transportation safety. Airport observations of visual range have been made since 1919, and have been computer archived since the late 1940's. Daylight observations involve viewing preselected visibility markers (large dark objects) at known distances from the observation point to determine the most distant marker that is visible.

Another traditional visibility metric is extinction coefficient, which is the attenuation of light per unit distance due to scattering and absorption by gases and particle in the atmosphere. Extinction coefficient is expressed in inverse length units (e.g., km<sup>-1</sup>) and is used primarily by scientists studying the causes of reduced visibility. Direct relationships exist between concentrations of atmospheric constituents and their contribution to extinction coefficient. Apportioning extinction coefficient to atmospheric constituents provides a method to estimate change in visibility caused by change in constituent concentrations. Calculation of the extinction coefficients from air quality models can be used to estimate the expected visibility changes from emission changes.

#### Visibility Impairing Particles

Light scattering, and to a lesser degree, light absorption by suspended particles are the most important contributors to visibility degradation. The influence of particles depends on the concentration, composition, and the size of the particles. Particles composed of materials such as

sulfates and nitrates absorb under high relative humidity conditions. Since the solution drops are larger than the dry particles, visibility impairment by sulfate and nitrate particles increases during high humidity conditions.

Sulfate, nitrate, and organic carbon are major contributors to visibility degradation. These particles begin as gaseous emissions and undergo chemical transformation in the atmosphere. These particles are mostly in a size range from about 0.1 to 1.0 microns diameter which scatters more light for the same mass concentration than smaller or larger particles. That is, the particles that scatter light most efficiently per unit mass are those of approximately the same size as wavelengths of visible light (0.4-0.7 microns). Coarse particles (i.e., those in the 2.5 to 10 micron size range) also impair visibility, although less efficiently than fine particles. Black carbon, primarily from incomplete combustion such as in diesel exhaust or wood smoke, is the principal cause of light absorption in the atmosphere. Sulfates and nitrates readily absorb water from the atmosphere and grow in size in a nonlinear fashion as relative humidity levels increase. Because humidity varies seasonally, the visibility impacts of the sulfate and nitrate particles also varies by season.

#### NO<sub>x</sub> Emissions

As described in the "Ozone" section of this document, NO is formed during combustion or any high temperature process involving air. The NO is converted to  $NO_2$  by  $O_3$  or other atmospheric oxidants. The NO<sub>2</sub> then participates in various reactions to form other substances, including  $O_3$  and PM.

#### Nitrate Particulates

Nitrate airborne particles can be produced by several mechanisms. One major mechanism of nitrate formation involves the gas-phase reaction of NO<sub>2</sub> with OH to produce nitric acid, HNO<sub>3</sub>. The gaseous nitric acid can react at airborne particle surfaces to form nitrate salts. For example, a particle containing calcium carbonate can neutralize the nitric acid to produce calcium nitrate; it follows that nitrate size distributions depend, in part, on the size distributions of particles on which they react (NRC, 1993). Ammonia gas (NH<sub>3</sub>) is important in the generation of sulfate and nitrate particles through the neutralization of sulfuric and nitric acid. When concentrations of NH<sub>3</sub> and HNO<sub>3</sub> are sufficiently high, ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) can be formed. Ammonium nitrate is often found in submicron particles in locations such as Denver or Los Angeles. However, little submicron ammonium nitrate is typically found in parts of the nation where ammonia concentrations are low and acid sulfate concentrations are high (NRC, 1993). (See additional discussion in the "Particulate Matter, Emissions Sources and Formation Processes of Particles" section of this document.)

#### $NO_2$

The only gas that absorbs visible light to any appreciable extent at concentrations expected in the atmosphere is  $NO_2$ .  $NO_2$  is a strong absorber of visible and ultraviolet light and can thereby contribute to haze. In addition,  $NO_2$  has a broad absorption band at the blue end of the spectrum;

consequently, when  $NO_2$  concentrations are high, the atmosphere has a distinct brownish color. However, because of its high reactivity and relatively short lifetime,  $NO_2$  does not normally contribute significantly to haze in remote areas; it is a problem only in areas close to sources (NRC, 1993).

The most significant optical effect of NO<sub>2</sub> involves discoloration (EPA, 1996). NO<sub>2</sub> appears as a yellow to reddish-brown gas because it strongly absorbs blue light, allowing red wavelengths to reach the eye. The extent to which NO<sub>2</sub> filters out blue light is determined by the integral of NO<sub>2</sub> concentration along the sight path. In regard to regional haze, because the effect of NO<sub>2</sub> depends on the product of the pollution concentration and the viewing path length, the coloration of 0.05 ppm NO<sub>2</sub> over 10 km is the same as 0.5 ppm over 1 km. When NO<sub>2</sub> is dispersed over a large area, as in the case of urban emissions, ground level concentrations at individual points may be less than a national standard but because an observer views the entire NO<sub>2</sub> mass, the urban plume appears of brownish color.

#### Significance of Anthropogenic Sources of Fine Particles

The concentrations of background fine particles are generally small when compared with concentrations of fine particles from anthropogenic sources. The same relationship holds true when one compares light extinction due to background fine particles with light extinction due to anthropogenic fine particles. Anthropogenic contributions account for about one-third of the average extinction coefficient in the rural West and more than 80 percent in the rural East (NAPAP, 1991). In the eastern United States, sulfates dominate fine particle concentrations, stemming from regional SO<sub>2</sub> emissions. In contrast, nitrate plays a small role in the East but is significant in areas of the West; for example, nitrates dominate the overall light extinction in the mountainous areas just outside Los Angeles, with most of the nitrate formation in this area coming from NO<sub>x</sub> emissions within the urban area (EPA, 1996).

#### Monitoring Data

While the amount of total light extinction varies significantly across the country, so does the mix of visibility-impairing pollutants from region to region. As described in Table II-4, IMPROVE monitoring data were used to establish annual apportionment of current aerosol components to the total visibility impairment for class I areas. This gives an indication of the relative contribution to visibility impairment due to nitrates in a variety of areas. Nitrates are the largest contributor to light extinction in National Parks and Wilderness Areas in Southern California.

REGION	Total Extinction	Aerosol Extinction	Sulfate	Nitrate	Organics	Elemental carbon	Soil and Coarse
ALASKA	25.4	15.4	6.7	0.7	4.6	0.5	2.6
APPALACHIAN	112.2	102.2	69.7	6.9	16.7	4.6	4.3
BOUNDARY WATERS	68.2	58.2	29.8	8.4	14.1	2.2	3.8
CASCADES	58.8	48.8	19.0	3.3	19.2	4.9	2.4
CENTRAL ROCKIES	28.1	18.8	5.8	1.3	6.1	1.3	3.6
CENTRAL CALIFORNIA COAST	56.3	46.3	15.4	12.1	10.6	2.7	5.6
COLORADO PLATEAU	27.1	17.1	6.0	1.4	4.7	1.5	3.5
FLORIDA	87.5	77.5	42.4	9.5	15.4	3.6	6.7
GREAT BASIN	23.4	13.4	3.4	0.9	4.6	0.6	4.0
HAWAII	53.2	43.2	31.5	1.0	5.0	0.7	5.1
NORTHEAST	71.3	61.3	38.3	5.1	11.0	4.0	2.9
NORTHERN GREAT PLAINS	39.7	29.7	13.1	3.3	7.3	1.4	4.7
NORTHERN ROCKIES	54.3	44.3	12.4	4.0	19.6	4.3	3.9
SIERRA NEVADA	33.4	24.4	5.7	3.6	8.1	2.5	3.4
SIERRA HUMBOLDT	28.0	18.0	4.4	1.4	7.7	1.8	2.7
SONORAN DESERT	31.3	21.3	8.1	1.3	5.5	1.8	4.5
SOUTHERN CALIFORNIA	63.5	53.5	7.7	23.8	9.7	4.8	7.5
WASHINGTON, D.C.	164.3	154.3	75.6	24.6	25.0	18.4	10.6
WEST TEXAS	36.7	26.7	12.2	1.4	5.7	1.5	5.9

# Table II-4. Annual Averages (March 1988-February 1991) of Reconstructed Light Extinction (Mm<sup>-1</sup>) for 19 Regions of the IMPROVE Network. (Source: IMPROVE, CIRA Report, Feb. 1993)

4. How much reduction is needed regionally; nationally?

While the answers to these questions are not clear at this time, it is clear that the type and

amount of emissions reductions needed varies from area to area. Visibility conditions vary regionally, as a function of background concentrations of fine particles, average relative humidity levels, and anthropogenic particle loadings, all of which are generally higher in the East than in the West. It is important to note that even in areas with relatively low concentrations of anthropogenic fine particles, such as the Colorado plateau, small increases in emissions can lead to significant decreases in visual range. This is one reason for the emphasis on protecting visual air quality in the highly valued national parks and wilderness areas in the Colorado plateau region. In areas with relatively higher fine particle concentrations, such as the Great Smoky Mountains National Park, it takes a greater reduction in ambient concentration to make an equivalent improvement in visual range.

Most visibility impairment in national parks and wilderness areas results from the transport by winds of emissions and secondary airborne particles over great distances (typically hundreds of kilometers). Consequently, visibility impairment is usually a regional problem, not a local one. Progress towards the national goal of remedying and preventing anthropogenic visibility impairment in Class I areas will require regional programs that operate over large geographic areas; strategies should be adopted that consider many sources simultaneously on a regional basis (NRC 1993). The outcome of the GCVTC report and EPA's subsequent rulemaking will help define the needed reductions to meet the national goals.

5. How much reduction will be achieved with current and projected programs?

Implementation of the CAA will achieve substantial decreases in  $NO_x$  emissions. The PSD program has managed atmospheric loadings from new sources and has safeguarded some large parklands from excessive emissions from new sources. Programs that may achieve additional  $NO_x$  emissions reductions are described in the Appendices to this document. Further, the revisions to the PM and  $O_3$  NAAQS could lead to additional emissions reductions.

6. Summary

Visibility impairment can occur due to local plumes or widespread regional haze. The sources of locally visible plumes are easy to identify, for example, the smoke from a power plant stack or from a burning field. However, when plumes are carried by winds, they become more diffuse, and the sources are identified less readily. In regions with many sources, the plumes can merge and become mixed with the emissions from many sources, such as motor vehicles, power plants, and industrial operations. The result is a widespread haze in which individual contributions from the various sources are virtually indistinguishable.

In most cases, visibility degradation is caused by five kinds of particulate substances: sulfates, nitrates, organic matter, elemental carbon, and soil dust. Regional haze is produced from a multitude of sources and impairs visibility in every direction over a large area, such as an urban area, or possibly over several states. Multiple sources may combine over many days to produce haze, which is often regional in scale. The fate of regional haze is a function of

meteorological and chemical processes, sometimes causing fine particle loadings to remain suspended in the atmosphere for long periods of time and to be transported long distances from their sources. The major cause of visibility impairment in the East is sulfate, formed primarily from  $SO_2$  emitted from coal combustion by electric utility boilers, while in the West the other four particle types play a greater role. Emissions of  $NO_x$  lead to the formation of particulate nitrates; thus, the reduction in emissions of  $NO_x$  will help improve visibility and make progress toward the national goal.

References

Grand Canyon Visibility Transport Commission, *Recommendations for Improving Western Vistas*, June 10, 1996 report of the Grand CanyonVisibility Transport Commission to the United States Environmental Protection Agency, June 10, 1996.

National Acid Precipitation Assessment Program (NAPAP) 1979 and the 1990 Reports *Acidic Deposition: State of Science and Technology*, Volume III, Report Number 24, entitled "Visibility: Existing and Historical Conditions - Causes and Effects."

National Acid Precipitation Assessment Program (NAPAP), (1991). Office of the Director, *Acid Deposition: State of Science and Technology*. Report 24, "Visibility: Existing and Historical Conditions - Causes and Effects." Washington, D.C.

National Research Council, Committee on Haze in National Parks and Wilderness Areas *Protecting Visibility in National Parks and Wilderness Areas*. National Academy Press, Washington, D.C., 1993.

Science and Technical Support Work Group, Ozone and Particulate Matter National Ambient Air Quality Standards, Federal Advisory Committee Act, Harvey Jeffries and Tom Helms, co-chairs, "Conceptual Model for Ozone, Particulate Matter and Regional Haze," February 19, 1997.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Effects of the 1990 Clean Air Act Amendments on Visibility in Class I Areas: An EPA Report to Congress. Research Triangle Park, N.C., 1993.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *Review of the national ambient air quality standards for particulate matter: policy assessment of scientific and technical information*, (Staff Paper), EPA-452/R-96-013, July 1996, NTIS # PB-97-115406.

III. Additional Public Health and Environmental Impacts from  $\mathrm{NO}_{\mathrm{x}}$  Emissions

A. Drinking Water

As described in the section on "Acid Deposition," nitrate and nitrite leaching aggravates the effects of acidification, both long term (e.g. cation leaching) and episodically (e.g. Al peaks). Adding inorganic N to freshwater ecosystems can eutrophy as well as acidify the waters when they are already rich in phosphorus or, more rarely, when N is the limiting nutrient (Vitousek et al, 1997:10; Church, 1997:17).

Most freshwater ecosystems will not be eutrophied by additional nitrate or nitrite leaching because phosphorous is the limiting nutrient. (Because the direct contribution of nitrite is typically very small compared to nitrate, sometimes this process is called simply "nitrate leaching.") Nitrate leaching has implications that go beyond its eutrophying and acidifying effects. Because primary producers (plants) in most freshwater systems do not assimilate added nitrate, this ion can remain in the water and be transported long distances downstream. By contrast, acidity may be neutralized by natural processes before it reaches large streams, lakes or estuaries. This characteristic leads to two impacts that can occur downstream: elevated levels of nitrate in drinking water supplies (discussed below) and eutrophication of estuaries and coastal waters (discussed in the following section). These downstream impacts can occur in hydrological systems even where acidification is not a problem.

Under the Safe Drinking Water Act, EPA has set maximum contaminant levels (MCLs) for nitrate and nitrite in drinking water to protect human health. These levels are established at 10 milligrams per liter (mg/L) of nitrate N and 1 mg/L of nitrite N (40 CFR 141.62). The primary adverse human health effect associated with exposure to nitrate or nitrite is methemoglobinemia (National Research Council, 1995:2). To cause methemoglobinemia, nitrate must be converted into nitrite. Methemoglobinemia occurs when nitrite oxidizes iron ( $Fe^{2+}$ ) in blood into  $FE^{3+}$ , a form that does not allow oxygen transport, and can cause brain damage or death (Vitousek et al, 1997:9). This condition in adults is rare (National Research Council, 1995:2), but is a significant concern for infants because microbes in an infant's stomach may convert high levels of nitrate to nitrite (Vitousek et al, 1997:9). Insufficient oxygenation of the blood is characterized by bluish skin and lips ("blue baby syndrome"). The National Research Council concluded, in 1995, that "results from epidemiological studies are inadequate to support an association between nitrate or nitrite exposure from drinking water in the United States and increased cancer rates in humans" (National Research Council, 1995:2). However, an epidemiological study, published in 1996, of rural populations using community water supplies in Nebraska, concluded that "long-term exposure to elevated nitrate levels in drinking water may contribute to the risk for Non-Hodgkin's Lymphoma (NHL)" (Ward et al, 1996:465).

The contribution of atmospheric N deposition to elevated levels of nitrate in drinking water supplies is an evolving impact area. The Ecological Society of America (ESA) has included discussion of this impact in a recent major review of causes and consequences of human alteration of the global N cycle in its *Issues in Ecology* series (Vitousek et al, 1997:9). This series is designed to report, in language understandable by non-scientists, the consensus of a panel of scientific experts on issues relevant to the environment. *Issues in Ecology* is supported by the Pew Scholars in Conservation Biology Program and ESA, a national professional society founded in 1915.

For decades, N concentrations in major rivers and drinking water supplies have been monitored in the United States, Europe, and other developed regions of the world. Monitoring data from public water systems in the United States show that surface water sources of drinking water do not exceed MCLs for nitrate and nitrite, with very rare exceptions unrelated to airborne deposition. On the other hand, analysis of these data confirms a substantial rise of N levels in surface waters, which are highly correlated with human-generated inputs of N to their watersheds. These N inputs are dominated by fertilizers and atmospheric deposition (Vitousek et al, 1997:9). Nitrate levels in the Mississippi River have more than doubled since 1965; they have risen in major rivers of the northeastern U.S. by three- to ten-fold since 1900 (Vitousek et al, 1997:9).

Agricultural sources dominate human-generated inputs of N in regions with intensive farming, including the Mississippi River basin and Texas. In other areas, such as the northeastern United States,  $NO_x$  emissions from industrial origin are the major factor in river export of N to lakes, major rivers, and estuaries (Howarth et al, 1996). Relatively low nitrate water from forested watersheds can serve to dilute higher nitrate water from urban or agricultural watersheds for many towns and cities. As water from forested watersheds increases in nitrate, therefore, its dilution capability diminishes and thus can threaten community water supplies even before actual nitrate concentrations exceed the drinking water standard (Ryan, 1996). Nitrate levels tend to be higher in private wells than community water supplies due to lack of regular testing, simpler construction, and shallow depth (Ward et al, 1996:165). High levels of nitrate in private well water typically indicate that pollution is seeping in from septic tanks, animal wastes, fertilizers, municipal landfills, or other nonpoint sources (EPA, 1996).

Increases in atmospheric N deposition to sensitive forested watersheds approaching N saturation would be expected to result in increased nitrate concentrations in stream water. Only one incidence of this phenomenon in the United States has been documented in peer-reviewed literature, although it has been well-established for areas in Germany and the Netherlands (Riggan et al, 1985:786). High nitrate concentrations from chaparral watersheds have been found in stream water in the South Coast (Los Angeles County, CA) Air Basin (Riggan et al, 1985, 1994). Stream water nitrate concentrations in watersheds subject to chronic air pollution were two to three orders of magnitude greater than in chaparral regions outside the air basin. Within the San Gabriel Mountains, nitrate concentrations were greatest, as high as 7.0 mg/L, where watersheds

adjoin the urbanized basin and may contribute to existing groundwater nitrate pollution (Riggan et al, 1985:786, 781).

Some high nitrate concentrations reported in this study were measured after a storm washed ash from burned landscape into the stream water. Wildfires in chaparral subject to chronic air pollution may cause inordinately high stream water nitrate loading which, when released through high stream discharge and channel scouring, could saturate the aquatic system and seriously pollute downstream waters (Riggan et al, 1985:788). These chaparral watersheds are specialized ecosystems and probably are not typical of United States watersheds in general. Nonetheless, high nitrate concentrations have also been observed in forest soils in the vicinity of the Asheville Watershed in western North Carolina; the amounts in soil solution suggest elevated levels of nitrate in groundwater (Smithson, 1997).

While observational evidence directly linking atmospheric deposition to elevated levels of nitrate in drinking water supplies is very limited, atmospheric deposition can supply N to ecosystems in a manner not dissimilar to fertilizer application (Vitousek et al, 1996:7-8). In fact, researchers have noted that some amount of atmospheric N deposition can be beneficial to agriculture, as fertilizer, in some areas, depending on the soil, crops harvested, biological uptake processes, and other factors (Acid Rain Program, 1995:36). Thus, one would expect that decreases in atmospheric N deposition to N-sensitive watersheds resulting from decreases in local or transported  $NO_x$  or ammonia N emissions would lessen the contribution of airborne deposition to elevated levels of nitrate in drinking water supplies.

# References

Acid Rain Program. 1995. Adverse Effects of Nitrogen Oxides ( $NO_x$ ) and Benefits of Reduction, draft staff assessment paper (Air Docket A-95-28, Item II-A-13). U. S. Environmental Protection Agency; Washington, DC. December.

Devesa, S.S. and T. Fears. 1992. "Non-Hodgkin's Lymphoma Time Trends: United States and International Data" *Cancer Res.* 1992; 52(suppl): 5432S-5449S.

Howarth, R.W., G.Billen, D.Swanwy, A. Townsend, N. Jaworski, K. Lajtha, J.A.Downing, R.Elmgren, N. Caraco, T. Joddan, F. Berendse, J. Freney, V. Kudeyarov, P. Murdoch, Zhu Zhao-Liang. 1996. "Regional nitrogen budgets and riverine N &P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences." *Biogeochemistry* (in press).

National Research Council. 1995. *Nitrate and Nitrite in Drinking Water*. Washington, DC: National Academy Press.

Riggan, P.J., R.N. Lockwood, and E.N. Lopez, "Deposition and Processing of Airborne Nitrogen Pollutants in Mediterranean-Type Ecosystems of Southern California" *Environmental Science and Technology*, vol. 19 (p. 781-789), 1985.

Riggan, P.J., R.N. Lockwood, P.M.Jacks, C.G.Colver, F.Weirich, L.F.DeBano, and J.A.Brass, "Effects of Fire Severity on Nitrate Mobilization in Watersheds Subject to Chronic Atmospheric Deposition" *Environmental Science and Technology*, vol. 28 (p. 369-375), 1994.

Ryan, Douglas F., USDA, Forest Service, Forest Environment Research, personal communication November 7, 1996.

Smithson, Paul. 1997. Unpublished doctoral dissertation, North Carolina State University, Department of Soil Science (p. 95-96, 115, 122, 130), 1997.

U.S. Environmental Protection Agency, "Environmental Indicators of Water Quality in the United States: Fact Sheets". EPA-841-F-96-001, 1996.

Vitousek, Peter M., John Aber, Robert W. Howarth, Gene E. Likens, et al. 1997. Human Alteration of the Global Nitrogen Cycle: Causes and Consequences. *Issues in Ecology*. Published by Ecological Society of America, Number 1, Spring 1997.

Ward, M.H., S.D. Mark, K.P. Cantor, D. D. Weisenburger, A. Correa-Villasenor, and S.H. Zahm. 1996. "Drinking Water Nitrate and the Risk of Non-Hodgkin's Lymphoma" *Epidemiology* 7:465-471.

#### B. Eutrophication

#### 1. Goals of the Program

Eutrophication has been identified as the most serious pollution problem facing the estuarine waters of the United States (NRC, 1993). In fact, 54 percent of impacted square miles of estuaries that have been assessed have been shown to be impaired by nutrients (EPA 1994). N is the limiting nutrient controlling eutrophication in most temperate estuaries (Nixon, 1986, Swain et al, 1994) and some limited percentage of freshwater lakes (EPA, 1993). Studies of N loadings to estuarine and coastal systems within the United States and worldwide have shown that N deposited from the atmosphere is a significant portion of the total N loadings, ranging from 10 to 70 percent, with 27 percent of the N loadings delivered to the Chesapeake Bay originating from atmospheric sources (Linker et al, 1993; Paerl. 1993, Valigura et al, 1995). N has been shown to be the nutrient controlling the baywide reductions of bottom water dissolved oxygen in Chesapeake Bay (Thomann et al. 1994).

The EPA Office of Air Quality Planning and Standards' program addressing the issue of N deposition and resultant eutrophication impacts is the Great Waters Program. The charge of the Great Waters program, under section 112(m) of the CAA amendments, is to evaluate the deposition of hazardous air pollutants (and other pollutants at the discretion of the Administrator) to the Great Lakes, Lake Champlain, Chesapeake Bay and other estuarine and coastal waters. Given the discretion for additional pollutants, and the lack of attention to the NO<sub>x</sub>/eutrophication link, EPA chose to use this program as a vehicle to address this N deposition/eutrophication issue. The Great Waters Program provides information in biennial reports to Congress regarding: the adverse effects of the deposition on human health and the environment; the proportion of the loading which comes from the atmosphere; the sources of the pollution; evaluations of the effectiveness of existing regulatory programs in addressing these problems; and, ultimately, what changes to regulations are needed to prevent the identified adverse effects. The overall goal of the Great Waters Program is to identify and prevent adverse effects due to air pollutants deposited to aquatic ecosystems.

The Great Waters program works cooperatively with a number of other EPA offices, the National Oceanic and Atmospheric Administration (NOAA), and state agencies with respect to the N deposition/eutrophication issue, including the Chesapeake Bay Program Office, the Office of Water, various National Estuary Programs (e.g., Tampa Bay), and NOAA's Air Resources Laboratory and Coastal Ocean Program Office. The goals of these programs are complementary to and intersect with the Great Waters goal of characterizing and addressing the water quality problems due to atmospheric deposition of N compounds.

2. Status of the Program

#### Great Waters Program

Because of established monitoring data bases, monitoring methodology research, enhanced monitoring networks, and linked airshed-watershed-water quality models available or under development, the Great Waters Program has been focused on further advancing efforts in specific geographic areas. For estuarine issues, that work has focused on the Chesapeake Bay. Participants in the Great Waters Program believe that providing a comprehensive evaluation in a well-studied geographic area provides an invaluable understanding of the total scope of the problem, causes, processes, and the range of feasible control/reduction/prevention management options -- an understanding that would be impossible to develop with a number of disparate, less-intensive studies. The Great Waters Program has already found that the information developed through the Chesapeake Bay Program is directly applicable to other east coast estuarine systems with adjustments needed for the respective waterbody's watershed characteristics. This direct transferability of technologies and technical findings will save similar financial investments and years of effort by the many other place-based estuarine and coastal management programs along the Atlantic and Gulf coastlines.

Since the First Report to Congress on the atmospheric deposition of pollutants to the Great Waters, studies of other coastal waters, at National Estuary Program waters in particular, have investigated the significance of atmospheric deposition of N compounds to their waters. To improve understanding and reduction of N deposition to Chesapeake Bay and other coastal waters, the Chesapeake Bay Program, various National Estuary Programs, and the Gulf of Mexico Program continue to develop and refine modeling and monitoring efforts by addressing uncertainties such as N retention in watersheds, the differences in transport and fate of various N compounds, and the contribution of near shore ocean waters to the N inputs to estuaries.

The second report to Congress on the atmospheric deposition of pollutants to the Great Waters was completed in June 1997 (EPA, 1997). The report continues to find N compounds a pollutant of concern. Atmospheric deposition of N compounds can contribute significantly to eutrophication in coastal waters, where plant productivity is usually limited by N availability. Accelerated eutrophication and its subsequent effects such as nuisance algal blooms and reduced oxygen levels pose significant problems for Chesapeake Bay and many other estuaries. The report also indicates that substantial progress has been made in addressing N contamination issues in Chesapeake Bay, the largest United States estuary. A strategy has been developed by the Chesapeake Bay Program for reducing the N load to the Bay. Part of this process includes the large-scale modeling and understanding of the type and geographic origin of airborne N to the Bay. Significant data also have been collected on rates and amounts of N deposition (including comparison of direct and indirect deposition and of wet and dry deposition), and models have been developed to evaluate the impact of several N reduction scenarios on the Bay's water quality.

#### Chesapeake Bay Program

The Chesapeake Bay, largest of the 130 estuaries in the United States, was the first in the nation to be targeted for restoration as an integrated watershed, airshed, and ecosystem. The 64,000 square mile drainage basin covers parts of six states including New York, Pennsylvania, Maryland, Delaware, West Virginia, and Virginia and includes more than 150 tributaries. Now

in its thirteenth year, the Chesapeake Bay Program is a unique, regional, federal-state-local partnership that has directed and coordinated Chesapeake Bay restoration since the signing of the historic 1983 Chesapeake Bay Agreement. The Chesapeake Bay Program includes the state of Maryland, the Commonwealths of Virginia and Pennsylvania, the District of Columbia, the Chesapeake Bay Commission (representing the three state legislatures), and EPA on behalf of more than 25 participating federal departments and agencies.

Building on an expanded understanding of the Bay system and increasing experience with on-the-ground implementation within the cooperative basinwide partnership, the 1987 Chesapeake Bay Agreement set forth a comprehensive array of goals, objectives and commitments addressing living resources, water quality, population growth and development, public information, and governance (Chesapeake Executive Council 1987). The centerpiece of the agreement was a commitment to achieve a 40 percent decrease of controllable N and phosphorus entering Chesapeake Bay by the year 2000. Atmospheric deposition was considered part of the uncontrollable portion of nutrient loadings to the Bay tidal waters under the 1987 Bay Agreement. Through amendments to the Bay Agreement, the signatories have since committed to "quantify the impacts and identify the sources of atmospheric inputs on the Bay system" and incorporate atmospheric deposition as an integral component of the tributary basin-specific nutrient decrease strategies (Chesapeake Executive Council 1992, 1993).

#### Airshed-Watershed Models

While the current Chesapeake Bay models can simulate the relative effects of atmospheric N deposition on water quality, the ultimate goal is to link airshed emissions directly to ecological responses within a single model simulation framework. To provide for this predictive capacity, the Chesapeake Bay Program is configuring the Bay Watershed Model to accept daily atmospheric loadings to land use categories--forest, pasture, cropland, and urban (Chesapeake Bay Program 1995). The model can then simulate increased or decreased atmospheric loadings to the Bay tidal waters along with nutrients from other land-based point and nonpoint sources. The estuary model is being upgraded to simulate basic ecosystem processes of submersed aquatic vegetation, benthic microorganisms, and major zooplankton groups. With these refinements, the overall loads of controllable and uncontrollable N from the surrounding airshed and watershed, and the impact of these loads on the ecosystem can be simulated and evaluated. In parallel, Chesapeake Bay Program state and federal managers are developing the tools and information necessary to assess the efficacy of atmospheric source control options for basin-wide and tributary nutrient decrease strategies. The integrated Bay airshed-watershed-estuary-ecosystem models will be completed in mid-1997.

#### N Deposition Workshops

The Chesapeake Bay Program sponsored a coastal shared resources initiative which focuses on airsheds, the coastal ocean, migratory waterfowl and neotropical birds, and migratory fish. As part of this initiative, two workshops have been held to address atmospheric N deposition to estuarine and coastal waters. The first workshop was held at the Airlie House in Warrenton, Virginia, in October 1995 (the "Airlie workshop"). There, leading scientists and key policy and regulatory officials assembled to explore mechanisms by which air and water

pollution control programs can work together to help protect eastern coastal ecosystems<sup>11</sup>. The focus of the workshop was on atmospheric N compounds, but many of the conclusions would apply equally well to other pollutants occurring in the air, such as toxic chemicals, trace metals, and persistent organic compounds. In all such instances, the atmosphere constitutes a resource that is shared among many different coastal jurisdictions, as well as between state and federal air and water regulatory agencies.

The workshop concluded that there is need for (1) a better understanding of how all atmospheric N species affect coastal ecosystems, and of the related policy options, (2) a crossmedia approach to the atmospheric deposition and loading problem, and (3) a coalition of interested parties extending from the north to the south of the potentially affected eastern coast of the continental United States, including terrestrial, atmospheric, and aquatic aspects as equals. The work that is needed is essentially multi-media, requiring attention by a consortium of workers. The workshop resulted in a call for more cooperation across different issues, estuaries and bays, scientific disciplines, and state and federal agencies. It was determined that outreach to state and federal agencies, non-government organizations, industry, and the public at large, is critically needed.

To structure this effort, an action plan is included as the final section of the workshop report. The focus of short term actions is on effectively conveying the importance of the N deposition issue to both public citizens and officials in a unified voice. Mid-term, the emphasis shifts to improving cooperation across disciplines, estuaries and coastal waters, and agencies. And finally, the long-term actions deal with applying cross-media practices of all environmental issues and concerns detailed in the workshop report (East Coast Atmospheric Resource Alliance, 1996). (A copy of the report may be obtained from the NOAA Chesapeake Bay Office.)

The second workshop was held in Raleigh, North Carolina in March, 1997 (the "Raleigh workshop"). There were three broad objectives for this workshop: (1) to determine the essential connections between issues, programs, agencies, organizations and jurisdictions which could advance the ability to address the atmospheric N issue; (2) to identify and/or create new platforms for discussion of solutions to these problems; and (3) to identify management issues around which additional research and policy work are needed to advance the understanding of the ecosystem impacts for both airsheds and watersheds. Many of the questions addressed at the Airlie workshop were carried forward to the Raleigh workshop. Participants at the Raleigh workshop also endorsed a list of practical studies developed and prioritized at an earlier workshop in 1994 (described below). A report on the proceedings of the Raleigh workshop will be made available.

<sup>&</sup>lt;sup>11</sup>This workshop was convened as part of a larger east coast alliance of national estuary programs and coastal management programs under the Chesapeake Bay Program sponsored coastal shared resources initiative. The shared resource issues of focus include airsheds, the coastal ocean, migratory waterfowl and neotropical birds, and migratory fish.

#### NOAA Air Resources Laboratory

The NOAA Air Resources Laboratory conducts work in collaboration with academic researchers and EPA and includes air and deposition monitoring, as well as modeling and modeling support. Relevant activities include the following.

I. Strategic Scientific Planning: In an effort to coordinate scientific programs to reduce existing uncertainties in atmospheric loadings estimates, NOAA Air Resources Laboratory, in coordination with the Chesapeake Bay Program convened a workshop in June 1994, inviting scientists and managers with expertise and experience in understanding or managing atmospheric deposition. The challenge given to the workshop was simple--to construct a list of practical studies that would make the greatest impact on reducing the current uncertainty in estimates of the contribution of atmospheric deposition to declining coastal aquatic ecosystem health. The listing that resulted is summarized below and substantiated in the workshop proceedings report *Atmospheric Loadings to Coastal Areas: Resolving Existing Uncertainties* (Chesapeake Bay Program, 1995).

- Priority 1 -- Conduct intensive, coordinated integrated monitoring at special locations within the watershed, with wet deposition, dry deposition, and local catchment area characterizations -- to provide quality integrated monitoring data for evaluating model performance.
- Priority 2 -- Work to improve existing atmospheric models -- to address limitations of current models, especially limited resolution and the inability to handle orographic and chemical factors that are likely to be of critical importance.
- Priority 3 -- Improve biogeochemical watershed models -- to characterize watershed chemical processing and retention better.
- Priority 4 -- Improve emissions inventories and projections -- to provide more accurate inputs for deposition models, with better spatial resolution.
- Priority 5 -- Conduct process-oriented measurements to extend vertical and spatial meteorological and chemical concentration and deposition coverage and to quantify representativeness -- to provide more advanced input data as models evolve and input data requirements increase.
- Priority 6 -- Establish an extensive array of less intensive measurements -- (this item follows from Priority 1) to provide a nested network with a small number of Priority 1 intensive stations supporting a denser array of simple stations designed to provide improved spatial resolution for some selected variables.

II. Dry Deposition Inferential Method Network (DDIM): Dry deposition is a component that is not well characterized, because it is difficult to measure and to emulate

deposition to water. Data from Wye, Md. have revealed deposition rates which were similar to those reported from other DDIM stations in the region, and that dry deposition of nitrate ( $HNO_3/NO_3$ ) was approximately 46 percent of total nitrate deposition. However, further analysis indicated that some of the data may be suspect due to artifacts associated with sampler design affected by high ammonia and sea salt concentrations over the Eastern Shore. Future field studies will work to determine the validity of using the DDIM system in coastal areas.

III. Nitrogen Photochemistry: A collaborative research effort with the University of Maryland focused on nitrogen photochemistry and transport of anthropogenic precursors from local and regional sources. Results from this project indicate that highest  $NO_x$  concentrations occur when the source air mass has passed over the industrial midwest and/or the Washington-Baltimore area. Furthermore, it appears that urban airshed models may underestimate  $NO_x$  concentrations in near-urban environments. These results need to be verified and reinforced with further studies.

IV. Precipitation Chemistry: A daily precipitation chemistry site was established in the lower Chesapeake Bay, on an island chosen to emulate precipitation deposition directly to a water body. The data from this site will allow (for the first time) the evaluation of deposition model outputs such as those given by RADM.

V. NO Off-gassing: Efforts are in process to investigate soil emissions of NO at Wye, MD.

VI. Meteorological Measurement: NOAA provided meteorological information support for the Atmospheric Exchange Over Lakes and Oceans experiment in August 1995. In addition, in late March 1992, the first Chesapeake Bay Observing System (CBOS) buoy was placed by the University of Maryland in the North Bay, Maryland, the first of a proposed system of up to 11 similar buoys to record physical measurements. Given certain assumptions, the meteorological measurements made from the buoy allow the estimation of dry deposition velocities for nitric acid to the Bay surface. These measurements are being used to evaluate the ability of mesoscale atmospheric models to calculate deposition rates for N chemicals.

VII. Modeling: Operational forecasts and hindcasts are being performed and archived twice per day with the Regional Atmospheric Modeling System (RAMS) model. Work is currently underway to compare the RAMS predictions for over-water physical parameters with that measured by the CBOS buoys, and to reduce the RAMS resolution to characterize the Baltimore plume better.

#### Other Estuary Programs

The Chesapeake information is serving as the base for work in the Tampa Bay, where the Great Waters Program has begun leveraged support for the Tampa Bay National Estuaries Program for  $NO_x$ , organic N, and ammonia deposition sampling, as well as the sampling needed

to characterize the linkage between atmospheric deposition and urban storm water runoff. This effort began in 1995 and is expected to be complete in 1998. Further work in estuaries will be targeted in cooperation with the EPA Office of Water and the National Estuary Programs.

# 3. Science of $NO_x$ and Eutrophication

Though the availability of N normally limits biological productivity in coastal waters, over-abundance of N is of concern in areas which have developed nutrient enrichment problems, known as eutrophication. In addition to increasing primary productivity, nutrient enrichment generally alters the normal ratios of N to phosphorus and to other elements, such as silicon and iron. This alteration may induce changes to phytoplankton community structure. Species which normally occur in low abundance may be favored, and in some cases, toxic and/or noxious algal blooms may result. For the New England coast, for example, the number of red and brown tides and shellfish problems from nuisance and toxic plankton blooms have increased over the past two decades. In coastal areas with poor or stratified circulation patterns (e.g., Chesapeake Bay, Long Island Sound, Albemarle-Pamlico Sound) the "overproduction" of algae tends to sink to the bottom and decay, using all (anoxia) or most (hypoxia) of the available oxygen in the process, causing fish kills and loss of critical habitat. In some cases, the increase in suspended matter due to overproduction decreases light infiltration or algae grows directly on submerged living organisms, in turn causing a loss of submersed aquatic vegetation and coral communities.

Atmospheric deposition of N species is recognized by all east-coast estuarine programs as either a significant contributor to estuarine eutrophication or a mechanism of possible concern (East Coast Atmospheric Resource Alliance, 1996). The region from which the atmospheric N pollution arises is much larger than the water surface that is potentially affected, and even much larger that the watershed that drains into it. The extent of "airsheds" are now starting to be recognized. The Chesapeake Bay airshed (defined as the source region for 75 percent of deposited emissions) extends upwind of and borders the water body itself, reaching to New York, Ontario, Ohio, Tennessee and South Carolina (Dennis 1996). Emissions from within the Chesapeake Bay airshed may also affect estuaries along the coast from the Carolinas to New York. Similarly, the airsheds of other estuaries will overlap. Thus, airsheds constitute an important "shared resource" which must be recognized. Reductions in emissions in airsheds benefit downwind N-sensitive ecosystems, and assessments of the benefit of such decreases must take all benefitting water bodies into account, and not just one single ecosystem that is especially favored.

The uncertainties in studies to date make it important that a better understanding be obtained of the processes that transport and deposit N to estuaries and coastal zones. Current estimates are that emissions of  $NO_x$  are the largest contributor to atmospheric N loads to coastal waters of eastern North America (40-60 percent), with ammonia (20-40 percent) and organic N (about 20 percent) also contributing significant amounts on an annual basis (Paerl, 1993, 1995). The relative contribution of atmospheric N deposition to total new N inputs to estuarine, coastal, and offshore waters around the world ranges from less than 10 percent up to 70 percent (Paerl, 1993, 1995; Valigura et al, 1995). Studies performed on several other major East Coast

estuaries from Albemarle-Pamlico Sound to the Gulf of Maine have provided atmospheric N loading estimates that range between 18 percent and 39 percent of the total N load (East Coast Atmospheric Resource Alliance, 1996).

The most recent estimate of a 27 percent contribution of atmospheric deposition to total N loadings to the Chesapeake Bay falls within the range reported for other estuaries (Linker et al, 1993). Relative atmospheric contributions to coastal seas and open oceans may be higher, ranging up to 60-70 percent. However, a key factor is the actual amount (or total load) of N being considered. For example, the atmospheric contribution to the open ocean may be 70 percent compared to 27 percent in Chesapeake Bay, but the amount deposited (kg/ha) is small relative to that delivered to the Chesapeake Bay system (Paerl, 1995). In the coming decades, the atmosphere will become an even more significant source of N loadings to the Chesapeake Bay and other east coast estuaries when anticipated increases in population and land development result in increases in mobile source and power plant emissions. (Fisher, et al., 1988; Pechan, 1991).

#### Seasonal Impacts

Simulated water quality responses to year round vs seasonal nutrient decreases were conducted as a 1992 reevaluation of the baywide Tributary Nutrient Reduction Strategy by the Chesapeake Bay Program and indicated the need for year round controls on phosphorus and N loadings (Thomann et al 1994). The seasonal nutrient reductions scenarios, largely focused on assessing seasonal vs. year round biological nutrient removal at wastewater treatment facilities, indicated that winter N inputs contribute to summer eutrophication events. Further work is planned in the 1997 timeframe to more specifically explore the effects of seasonal controls from air emission sources.

#### 4. Needed Reductions - Regionally, Nationally

Obviously, characterizing the reduction of emissions needed to eliminate or ameliorate the atmospheric contribution to eutrophication of the nation's estuarine and coastal waters depends on a number of factors. These include: (1) amount of N in an estuarine/coastal ecosystem which constitutes a problem (i.e., the threshold at which nutrient inputs lead to eutrophication-related impacts); (2) relative contribution of specific atmospheric N compounds ( $NO_x$ ,  $NH_4^+$ , Organic N) to the total N loading and biotic impacts on the individual ecosystem; and (3) contribution of emission sources, or source categories, to deposition in specific estuarine or coastal waters.

The specific answers to these questions are going to differ for any given estuary, due to conditions of trophic status, hydrology, non-atmospheric N (and atmospheric ammonia and organic N) inputs, location and meteorology relative to  $NO_x$  and other N sources, development within the surrounding watershed, and other variables. Responses to these questions are being pursued by the EPA and NOAA and, to a growing degree, by Atlantic and Gulf national estuary programs as a result of the 1995 Shared Resources Workshop described previously. There are no complete answers yet, but insights into all three questions are emerging from work on

Chesapeake Bay. Some Bay efforts to provide information are described below.

#### Emission Source Characterization

From the major Bay influencing states--Maryland, Virginia, Pennsylvania, West Virginia, Ohio, New York, and New Jersey--utility sources (e.g., power plants) contribute 37 percent of the total  $NO_x$  emissions, mobile sources (e.g., cars, trucks) contribute 35 percent, other area sources (e.g., ships, boats, lawn equipment) contribute 21 percent, and other point sources (e.g., industries) contribute 6 percent (Dennis, 1996). These source emissions contributions are roughly equivalent to the source contributions to deposited ions of N from utility and mobile sources to the Chesapeake Bay and its watershed.

#### Atmospheric Deposition Modeling

Most assessments involving atmospheric deposition to the Chesapeake Bay have made use of the Regional Acid Deposition Model (RADM) (Dennis, 1995), which contains advanced descriptions of atmospheric chemistry and state-of-the-art formulations of atmospheric transport and dispersion. The model has been tested in considerable detail (Dennis, 1995). In practice, RADM is used in conjunction with the most advanced large scale watershed model available to yield not only estimates of deposition to the Chesapeake Bay watershed but also loadings to the Chesapeake Bay tidal waters.

#### Arced Delineation

Bounded by a set of decision rules, a series of scenarios run on the RADM were used to delineate the airshed of the Chesapeake Bay. The Chesapeake Bay airshed, roughly 350,000 square miles in size or more than 5 and a half times larger than the watershed, includes all of Maryland, Virginia, Pennsylvania, Delaware, the District of Columbia, West Virginia, and Ohio, most of New York, half of New Jersey, North Carolina, and Kentucky, and parts of Tennessee, South Carolina, Michigan, Ontario, and Quebec, including Lake Ontario and Lake Erie. The airshed is defined as that contiguous region of air emissions that contributes the majority of the deposition (75 percent) to the Bay and its watershed.

As defined, the Chesapeake Bay airshed, containing 30 percent of Eastern United States and Canadian emissions, accounts for 75 percent of the atmospheric N deposited onto the Bay and its watershed. The remaining 25 percent originates from emission sources outside the airshed. These remaining sources were located beyond a predefined point of diminishing return; that is, when a 50 percent reduction in emissions from large source regions would be expected to produce less than a 10 percent reduction in deposition onto the Bay watershed (Dennis, 1996). Therefore, the areal extent of the Chesapeake Bay airshed as defined here is an underestimate of the actual areas of the United States and Canada that contain sources that contribute to N deposition to the Bay and its watershed. A still undefined portion of the airshed is that portion which contributes to the atmospheric N deposition on coastal waters which, in turn, contributes to the influx of N from coastal waters into the southern Chesapeake Bay (Chesapeake Bay Program, 1994).

#### Relative Source Emission Contributions to Loadings

The pattern of N deposition from these difference sources were simulated using the RADM. These model simulations suggest that N emissions from utilities contribute a majority of the N which deposits on the western side of the Bay watershed, with a decreasing trend from the western to eastern portion of the watershed. The RADM runs further suggest that mobile sources (associated with the Boston-Washington megalopolis) contribute a majority of the N which deposits along the Delmarva Peninsula, the mainstem Chesapeake Bay, and the lower portions of the western shore tidal tributaries, with a decreasing trend from the eastern to western portion of the basin.

#### 5. Reductions with Current and Projected Programs

Reduction scenarios are part of an effort to determine how to achieve the goal of a 40 percent reduction in nutrient loadings to the Chesapeake Bay by 2000 (from a 1985 baseline). Land-based nonpoint source and wastewater treatment facility-based point source reduction actions, planned for implementation in many Chesapeake Bay tributary watersheds, are approaching the limits of technology. Options for reductions in air emissions are being explored for maintaining the targeted 60 percent nutrient loadings cap beyond the year 2000 in the face of a growing population and resultant development in the watershed.

Anticipated reductions in  $NO_x$  emissions, and resultant N loadings to estuarine waters, due to a range of CAA and Ozone Transport Commission regulatory and non-regulatory actions are currently being simulated through the linked Chesapeake Bay airshed-watershed-estuarine water quality ecosystem processes models. Present model estimates are that over one quarter of the total N loading to the Bay system comes from the atmosphere. However, significant refinements to the Bay Watershed Model are being conducted and final results are expected in mid-1997. Table III-1 shows the estimated changes in that loading that would result from two scenarios of  $NO_x$  control -- first, full implementation of the CAA amendments of 1990<sup>12</sup>, and second, implementation of the more intensive controls advocated by the Ozone Transport Commission<sup>13</sup>.

Table III-1. Estimated Reductions in N Loadings to Chesapeake Bay and Water Quality Response Under Several Control Scenarios.

<sup>&</sup>lt;sup>12</sup>The Clean Air Act Scenario estimates the atmospheric load reductions expected under full implementation of the Clear Air Act titles I, II, and IV.

 $<sup>^{13}</sup>$ The Ozone Transport Commission Scenario evaluates the effects of conditions found in Clean Air Act titles I, II, and IV, as well as additional N decreases to lower ground level O<sub>3</sub> in mid-Atlantic and New England metropolitan regions as called for by the Ozone Transport Commission.

Control Scenario	Nitrogen Atmo- spheric Deposition Reduction to Bay	Nitrogen Atmospheric Deposition Reduction to Watershed	Nitrogen Load Reduction to Bay (millions of lbs per year)	Water Quality Improvement*
Bay Agreement (no air controls)	0%	0%	75 (target)	20%
Clean Air Act	9%	14%	14	4% **
Ozone Transport Commission	12%	21%	24	6% ***

\* The water quality improvements are quantified in terms of estimated reductions in the volume of Bay bottom with no dissolved oxygen (i.e., reduction in Chesapeake Bay "dead waters"); decreased nitrogen loadings will also result in decreased water column nitrogen concentrations which will, in turn, decrease algae growth and so improve light penetration, necessary to support the critically important underwater Bay grasses (Batiuk et al., 1992; Chesapeake Bay Program, 1994d; Dennison et al., 1993; Thomann et al, 1994).

\*\* Determined by the difference between the Bay Agreement scenario alone and the Bay Agreement plus Clean Air Act scenario, *Response of the Chesapeake Bay Water Quality Model to Loading Scenarios*, Thomann, et al. 1994.

\*\*\* Extrapolation of Bay Agreement plus CAA scenario to load reductions under Ozone Transport Commission controls.

#### 6. Summary

One of the goals of the Great Waters program is to evaluate, and determine an effective means to address, the impacts of atmospheric N deposition to estuarine, and other N-limited waters. This goal is shared with the Chesapeake Bay Program, as well as many of the eastern National Estuary Programs. The efforts of these programs have been focused to date largely on the Chesapeake, using a variety of state-of-the-art models and monitoring data to evaluate the airshed, source categories of emissions, and ecosystem impacts. Work thus far has shown the relative contribution of the atmosphere to total N loadings to be significant. The airshed of the Chesapeake reaches from New York to Ontario, Kentucky, and South Carolina. Other affected estuaries are going to have similar, and overlapping airsheds.

The impacts of excess N, in these N-limited systems, is contributing to the most serious water quality problem in eastern estuaries at the present, eutrophication. The impacts of

eutrophication range from increased turbidity shading out beneficial submersed aquatic vegetation habitats, to the exacerbation of noxious algae blooms, to the creation of low or no-oxygen conditions (hypoxia or anoxia) which negatively affect fish populations.

In the Chesapeake, the affected parties have agreed that a 40 percent reduction in N inputs is needed for the Bay's health. The implementation of CAA provisions could account for about a fifth to a third of that. These same actions would be expected to provide similar benefits to yet unstudied estuaries with eutrophication problems.

#### References

Chesapeake Bay Program. 1995. *Atmospheric Loadings to Coastal Areas: Resolving Existing Uncertainties*. A report of the Atmospheric Loadings Workshop, Baltimore, Maryland June 29-30, 1994, organized and lead by the Air Quality Coordination Group and the Scientific and Technical Advisory Committee of the Chesapeake Bay Program.

Chesapeake Executive Council. 1987. 1987 Chesapeake Bay Agreement. Annapolis, Maryland.

Chesapeake Executive Council. 1992. Chesapeake Bay Agreement 1992 Amendments. Annapolis, Maryland.

Chesapeake Executive Council. 1993. Directive 93-1 Joint Tributary Strategy Statement. Annapolis, Maryland.

Dennis, R.L. 1996. Using the Regional Acid Deposition Model to determine the nitrogen deposition airshed of the Chesapeake Bay watershed. In: J.E. Baker (ed.). *Atmospheric Deposition of Contaminants to the Great Lakes and Coastal Waters*. SETAC Press, Pensacola, Florida.

East Coast Atmospheric Resource Alliance. 1996. Airsheds and Watersheds - the Role of Atmospheric Nitrogen Deposition. A report of the Shared Resources Workshop, October 11-12, 1995, Warrenton, Virginia.

Fisher, D.C., J. Ceraso, T. Mathew, and M. Oppenheimer. 1988. Polluted coastal waters: the role of acid rain. Environmental Defense Fund, New York, NewYork.

Linker, L.C., R.L.Dennis, and D.Y.Alegre. 1993. *Inpact of the Clean Air Act on Chesapeake Bay Water Quality*. International Conference on Environmental Management of Enclosed Coastal Seas. Baltimore, MD.

National Research Council. 1993. Report of the Committee on Wastewater Management for Coastal Urban Areas, Water, Science, and Technology Board. Washington, D.C.

Nixon, S.W. 1986. "Nutrient Dynamics and the Productivity of Marine Coastal Waters". R.

Halwagy, D. Clayton, and M. Behbehani (eds): *Coastal Eutrophication*. pp 97-115. The Alden Press, Oxford.

Paerl, H.W. 1993. "The Emerging Role of Atmospheric Nitrogen Deposition in Coastal Eutrophication: A Biogeochemical and Trophic Perspective." *Canadian Journal of Fisheries and Aquatic Sciences*. 50:2254-2269.

Paerl, H.W. 1995. "Coastal Eutrophication in Relation to Atmospheric Nitrogen Deposition: Current Perspectives." *Ophelia*. 41:237-259.

Pechan, E.H. 1991. Background documentation on  $SO_2$  and  $NO_x$  forecasts. E.H. Pechan and Associates, Springfield, VA. EPA Contract No. 68-D00120.

Swain, W., T. Colborn, C. Bason, R. Howarth, L. Lamey, B. Palmer, and D. Swackhamer. 1994. *Exposure and Effects of Airborne Contamination*. U.S. Environmental Protection Agency. EPA-453/R-94-085.

Thomann, R.V., J.R. Collier, A. Butt, E. Casman, and L.C. Linker. 1994. *Response of the Chesapeake Bay Water Quality Model to Loading Scenarios*. Chesapeake Bay Program Office, Annapolis, Maryland. CBP/TRS 101/94.

U.S. Environmental Protection Agency. 1994. National Water Quality Inventory: 1992 Report to Congress. Washington, D.C.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, revised criteria document, *Air Quality Criteria for Oxides of Nitrogen* (three volumes, EPA-600/8-91/049aF-cF, August 1993.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *Deposition of Air Pollutants to the Great Waters*, Second Report to Congress, EPA-453/R-97-011, June 1997.

Valigura, R.A., J.E. Baker, J. R. Scudlark, and L.L. McConnell. 1995. Atmospheric Deposition of Nitrogen and Contaminants to Chesapeake Bay and its Watershed. In: *Perspectives on Chesapeake Bay, 1994: Advances in Estuarine Science*. S. Nelson and P. Elliott, eds. Chesapeake Research Consortium, Edgewater, Maryland.

# C. Global Warming

#### Global Warming

The "greenhouse effect" is the name for the physical process whereby energy from the sun passes through the atmosphere relatively freely, while heat radiating from the earth is partially blocked or absorbed by water vapor and other radiatively important gases in the atmosphere. Because the sun is hotter than the earth, its energy is radiated at a higher frequency which is not absorbed well by gases such as carbon dioxide or water vapor. In contrast, these types of gases are effective absorbers of the lower-frequency energy radiated by the earth. Since the "greenhouse" gases responsible for this selective absorption make up only about one percent of the atmosphere, they are also known as "trace" gases. The energy absorbed by the different trace gases can be calculated accurately. When the concentration of a trace gas increases, this additional absorption warms the planet, if there are no other changes in the climate system.

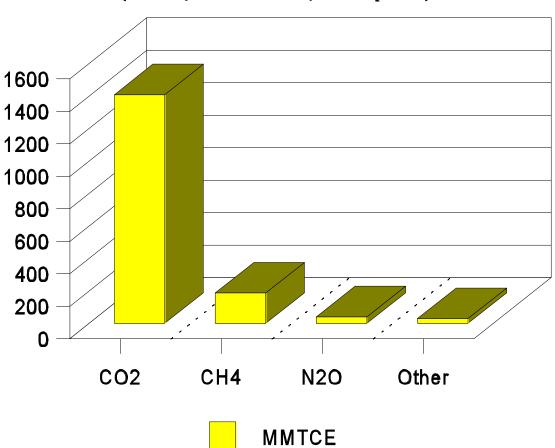
#### Greenhouse Gases

Naturally occurring greenhouse gases include water vapor, carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), and O<sub>3</sub> (EPA, November 1995). In addition, other photochemically important gases--such as carbon monoxide, NO, NO<sub>2</sub> and VOCs--are not greenhouse gases, but contribute indirectly to the greenhouse effect because they influence the rate at which O<sub>3</sub> and other gases are created and destroyed in the atmosphere. That is, emissions of NO<sub>x</sub> lead to the formation of tropospheric O<sub>3</sub>, which is also a greenhouse gas. As described below, some important sources of NO<sub>x</sub> are also emitters of N<sub>2</sub>O. Since 1800, atmospheric concentrations of carbon dioxide have increased by more than 25 percent, methane concentrations have doubled, and N<sub>2</sub>O concentrations have risen approximately 8 percent; this recent atmospheric buildup appears to be largely the result of anthropogenic activities (EPA, October 1995).

Figure III-1 illustrates the relative contribution of the greenhouse gases to total United States anthropogenic emissions in 1994 (EPA, November 1995). Due largely to fossil fuel consumption, carbon dioxide emissions accounted for the largest share of United States emissions on a carbon equivalent basis<sup>14</sup>--almost 85 percent. Methane emissions accounted for 11 percent and N<sub>2</sub>O emissions comprise about 2 percent of the global warming potential of all the United States greenhouse gases (EPA, November 1995). On a global basis,N<sub>2</sub>O is estimated to contribute 6 percent of the total global warming atmospheric gases (Kramlich and Linak, 1994).

<sup>&</sup>lt;sup>14</sup>In order to compare the ability of a greenhouse gas over time to trap heat in the atmosphere relative to another gas, the concept of global warming potential (GWP) is used. The GWP uses carbon dioxide as the reference gas with a GWP of 1. Thus, emissions of greenhouse gases may be reported in terms of million metric tons of carbon equivalent over a century. N<sub>2</sub>O, with a GWP of 320, has a much larger ability to trap heat than carbon dioxide on a gram for gram basis.





(U.S. 1994, Million Metric Tons, Carbon Equivalent)

#### $N_2O$

 $N_2O$  is a chemically and radiatively active greenhouse gas that is produced naturally from a wide variety of biological sources in soil and water, as well as from various anthropogenic sources. With an atmospheric lifetime of 150 years,  $N_2O$  is extremely long-lived and very stable in the troposphere (Kramlich and Linak, 1994). While actual emissions of  $N_2O$  are much smaller than carbon dioxide emissions,  $N_2O$  is approximately 320 times more powerful than carbon dioxide at trapping heat in the atmosphere over a 100-year time horizon (EPA, November 1995).

#### N<sub>2</sub>O Anthropogenic Emissions Sources

 $N_2O$  is produced naturally in soils through the microbial processes of denitrification and nitrification<sup>15</sup>. A number of anthropogenic activities add N to soils, thereby increasing the amount of N available for nitrification and denitrification, and ultimately the amount of  $N_2O$  emitted. Fertilizer use accounts for approximately 45 percent of total United States emissions of  $N_2O$ . Anthropogenic emissions of  $N_2O$  in the United States have increased over 10 percent from 1990 to 1994 primarily for two reasons: increased fertilizer use and general economic growth (EPA, October 1995). The major United States anthropogenic emissions sources are summarized in the Figure III-2 (EPA, November 1995).

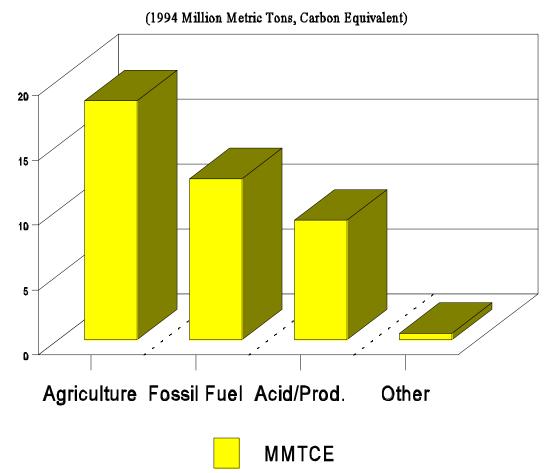
Other important anthropogenic activities producing N<sub>2</sub>O are fossil fuel combustion from mobile and stationary sources, adipic acid production, and nitric acid production, which contribute 31, 13, and 10 percent, respectively, of the total United States emissions of N<sub>2</sub>O. N<sub>2</sub>O is a product of the reaction that occurs between nitrogen and oxygen during fossil fuel combustion. Total emissions are estimated at approximately one half million tons or 41 million metric tons carbon equivalent (MMTCE)<sup>16</sup> (EPA, November 1995). Mobile emissions totaled 9.3 MMTCE in 1994, with road transport accounting for approximately 95 percent of these N<sub>2</sub>O emissions. N<sub>2</sub>O emissions from stationary fossil fuel combustion sources were 3.2 MMTCE in 1994. Also with respect to 1994, the production of adipic acid (used to produce nylon) generated 5.4 MMTCE of N<sub>2</sub>O. There are currently four plants in the United States that produce adipic acid. Since 1990, two of the plants have employed emissions control measures destroying about 98 percent of the N<sub>2</sub>O before its release into the atmosphere. By 1996, all adipic acid production plants will have N<sub>2</sub>O emissions controls in place as a result of a voluntary agreement among producers (EPA, November 1995). Production of nitric acid is another industrial source of N<sub>2</sub>O emissions. Nitric acid is a raw material used primarily to make synthetic commercial fertilizer; N<sub>2</sub>O emissions from this source were about 3.8 MMTCE in 1994.

<sup>&</sup>lt;sup>15</sup>Denitrification is the process by which nitrates or nitrites are reduced by bacteria, which results in the escape of nitrogen into the air. Nitrification is the process by which bacteria and other microorganisms oxidize ammonium salts to nitrites, and further oxidize nitrites to nitrates.

<sup>&</sup>lt;sup>16</sup>MMTCE is a method of comparing the global warming potential of various greenhouse gases. Carbon dioxide was chosen as the reference greenhouse gas.

Figure III-2

# Nitrous Oxide Emission Sources



# N<sub>2</sub>O Global Emissions Sources

 $N_2O$  is emitted by several sources, which have large uncertainties, and its global atmospheric budget is difficult to reconcile. The updated global budget is presented in Table III-2 below, expressed in million metric tons of N (WMO, 1994). Since the mid-1970s, systematic tropospheric measurements of  $N_2O$  have been made at locations worldwide. These data show the atmospheric concentration of  $N_2O$  to be increasing at an average rate of approximately 0.3 percent per year (Khalil and Rasmussen, 1992; Vitousek et al, 1997).

<u>Natural</u> 1.4-5.2 Oceans Tropical Soils Wet Forests 2.2-3.7 Dry Savannas 0.5-2.0 Temperate Soils Forests 0.5-2.0 ? Grasslands Anthropogenic Cultivated Soils 1-3 Animal Waste 0.2-0.5 0.2-1.0 **Biomass Burning** 0.1-0.3 Stationary Combustion Mobile Sources 0.1-0.6 0.5-0.9 Acid Production

TABLE III-2. Estimated global sources of N<sub>2</sub>O (million metric tons N per year)

#### $O_3$

 $O_3$  is an important greenhouse gas present in both the stratosphere and troposphere. Although representing only 10 percent of the total  $O_3$  column, tropospheric  $O_3$  is important because it can influence climate, as it is a greenhouse gas itself, and because its photolysis by UV radiation in the presence of water vapor is the primary source for hydroxyl radicals (WMO, 1994). Hydroxyl radicals are responsible for the oxidative removal of many trace gases, such as methane, hydrofluorocarbons, and hydrocholorfluorocarbons that influence climate and/or are important for the stratospheric  $O_3$  layer.

Observations show that free tropospheric  $O_3$  has increased above many locations in the Northern Hemisphere over the last 30 years. Model simulations and the limited observations together suggest that tropospheric  $O_3$  may have doubled in the Northern Hemisphere since pre-industrial times. Such changes in  $O_3$  have potentially important consequences for warming, although detailed quantification is not possible due to uncertainties in the size and distribution of the  $O_3$  change (Houghton, 1994).

Emissions of  $NO_x$  lead to the formation of tropospheric  $O_3$ . The increases in tropospheric  $O_3$  will be regional in nature and so will the associated effects noted above. Because changes in the tropospheric  $O_3$  are highly spatially variable, both regionally and vertically, assessment of global long-term trends is extremely difficult. To the extent reductions in emissions of  $NO_x$  would lower tropospheric  $O_3$  concentrations, the global warming effects of tropospheric  $O_3$  would also be lowered.

#### **Emissions Control Programs**

While the goal of many NO<sub>x</sub> control procedures for mobile and stationary sources is to convert NO into N<sub>2</sub>, some NO<sub>x</sub> control programs strive to prevent NO<sub>x</sub> emissions in the first place. For example, efforts to lower miles traveled by vehicles or agricultural methods to decrease the amount of N fertilizer applied will decrease both NO<sub>x</sub> and N<sub>2</sub>O. Other control programs are directed at N<sub>2</sub>O emissions, such as in adipic acid production. However, many NO<sub>x</sub> control procedures for mobile and stationary sources convert NO into N<sub>2</sub> and, in that process, a portion of the reactions will form N<sub>2</sub>O. Therefore, the development of an emissions control program to achieve a specific environmental goal may need to take into account the impact on both NO<sub>x</sub> and N<sub>2</sub>O emissions and the resultant environmental impacts that may go beyond the specific goal.

The amount of  $N_2O$  emitted is generally small compared to the  $NO_x$  emissions reductions and varies, depending upon fuel, technology type, and pollution control device. Emissions also vary with the size and vintage of the combustion technology, as well as maintenance and operation practices. Staged combustion, including low- $NO_x$  burners, and reburn technologies have only a small influence on  $N_2O$ . In selective non-catalytic reduction, use of  $NH_3$  results in less than 5 percent of the NO reduced being converted to  $N_2O$ ; use of urea converts more than 10 percent of the NO. The application of selective catalytic reduction suggests that  $N_2O$  emissions are negligible from vanadium catalysts; noble metal catalysts, however, may convert significant quantities of NO into  $N_2O$  (Kramlich and Linak, 1994). Regarding mobile sources, as catalytic converter-equipped vehicles have increased in the United States motor vehicle fleet, emissions of  $N_2O$  from this source have also increased (EPA, 1995).

References:

Houghton, J. T., et al, *Climate Change 1994*, "Radiative Forcing of Climate Change," reports of working groups I and III of the Intergovernmental Panel on Climate Change, Cambridge University Press, section 6.2.

Khalil, M.A. and Rasmussen, R.A., "The Global Sources of Nitrous Oxide," J. Geophys. Res. 97, 14651-14660, 1992.

Kramlich, J.C. and Linak, W.P, "Nitrous Oxide Behavior in the Atmosphere, and in Combustion and Industrial Systems," *Prog. Energy Combust. Sci.*, Vol 20, 1994.

U.S. Environmental Protection Agency, Office of Policy, Planning and Evaluation, *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-1994*, November 1995, EPA-230-R-96-006.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *National Air Pollutant Emission Trends*, 1900-1994, October 1995, EPA-454/R-95-011.

usek, P.M., J. Aber, R.W. Howarth, G.E. Likens, P.A. Matson, D.W. Schindler, W.H. Schlesinger, and G.D. Tilman, "Human Alteration of the Global Nitrogen Cycle: Causes and Consequences," *Issues in Ecology*, Number 1, Spring 1997.

World Meteorological Organization, *Scientific Assessment of Ozone Depletion: 1994*, Global Ozone Research and Monitoring Project--Report No. 37.

#### D. Stratospheric Ozone Depletion

# Stratospheric $O_3$

 $O_3$  is mainly found in two regions of the Earth's atmosphere. Over 90 percent of atmospheric  $O_3$  resides in the stratosphere, a layer between approximately 15 and 50 km (about 10-30 miles) above the Earth's surface (Kramlich and Linak, 1994). This stratospheric  $O_3$  is commonly known as the "ozone layer." The remaining  $O_3$  is in the lower region of the atmosphere, the troposphere, which extends from the Earth's surface up to about 10 km. Concentrations of  $O_3$  in the stratosphere are maintained by the balance between photochemical production of  $O_3$  (by photolysis of molecular oxygen,  $O_2$ ) and destruction of  $O_3$ . Stratospheric  $O_3$  is produced by the photolysis of  $O_2$ , which gives oxygen atoms (O), which then react with  $O_2$  to form  $O_3$ . Photochemical reactions associated with hydrogen oxides (HO<sub>x</sub>), NO<sub>x</sub>, and halogen oxides (ClO<sub>x</sub> and BrO<sub>x</sub>) destroy  $O_3$ . The relative contribution of each to  $O_3$  destruction varies with such factors as altitude, latitude, and season (WMO, 1994).

Stratospheric  $O_3$  plays a beneficial role by absorbing most of the biologically damaging ultraviolet sunlight called UV-B, allowing only a small amount to reach the Earth's surface. Many experimental studies of plants and animals, and clinical studies of humans, have shown the harmful effects of excessive exposure to UV-B radiation. In contrast, at the planet's surface, high concentrations of  $O_3$  are toxic to living systems and can damage the tissues of plants and animals. The ground-level  $O_3$  concentrations in the smoggiest cities are very much smaller than the concentrations routinely found in the stratosphere (WMO, 1994).

### Stratospheric O<sub>3</sub> Depletion

Decreases in stratospheric  $O_3$  have occurred since the 1970s. The most obvious feature is the annual appearance of the Antarctic  $O_3$  hole in September and October. The October average total  $O_3$  values over Antarctica are 50-70 percent lower than those observed in the 1960s; this phenomenon has come to be known as the Antarctic "ozone hole." The  $O_3$  loss occurs at altitudes between about 14 and 24 km. Smaller, but still significant losses in global total-column  $O_3$  have also been observed in the more populated mid-latitudes (30-60 degrees) of both hemispheres (WMO, 1994). For example, in the middle latitudes of the Northern Hemisphere, downward trends of about 6 percent per decade over 1979-1994 were observed in winter and spring and about 3 percent per decade were observed in summer and fall.

#### Polar Regions

The principal cause of  $O_3$  loss in the polar regions is photochemistry involving the halogen species, chlorine and bromine (WMO, 1994). Long-lived halogen species, primarily chlorofluorocarbons, are released in the troposphere from human activities. The photochemical degradation of these organic source molecules in the stratosphere leads to the formation of

inorganic halogen species. The release of chlorine from these species occurs in high-latitude winter in reactions on surfaces of stratospheric aerosol particles. The formation and reactivity of these particles are enhanced at the low temperatures characteristic of the interior of the polar vortices. The removal of reactive nitrogen, especially nitric acid, by aerosol particle sedimentation in the vortex strongly regulates the rate of recovery by controlling the availability of active chlorine. In the Antarctic, weather patterns result in the annual polar vortex, which prevents the transport of  $O_3$  from the southern hemisphere mid-latitudes to the polar regions, primarily in September and October.

#### Mid-latitudes

As noted above, stratospheric  $O_3$  concentrations are lowered by photochemical reactions associated with the hydrogen, N, chlorine and bromine radicals. The concentrations of these radical species are maintained by photodegredation of the corresponding source gases:  $H_2O$  and methane for  $HO_x$ ,  $N_2O$  for  $NO_x$ , and halogen source gases for  $ClO_x$  and  $BrO_x$ . Increases in radical concentrations (e.g., increases in  $ClO_x$  due to chloroflurocarbons emitted at the Earth's surface, and increases in  $NO_x$  due to  $N_2O$  emitted at the ground and stratospheric injection of  $NO_x$  by aircraft) lead to changes in  $O_3$  (WMO, 1994).

In the low stratosphere (10-22 km), reactions involving HO<sub>x</sub> dominate the O<sub>3</sub> loss rate, while between 23 and 40 km, NO<sub>x</sub> cycles dominate (WMO, 1994). Decreases in NO<sub>x</sub> above about 22 km, where it represents the dominant photochemical loss mechanism, would result in local stratospheric O<sub>3</sub> increases (Tie et al, 1994). The broad picture is of reactions involving HO<sub>2</sub> being responsible for over half the photochemical destruction of O<sub>3</sub> in the low stratosphere at midlatitudes, while halogen (chlorine and bromine) chemistry accounts for a further third. Although catalytic destruction by NO<sub>x</sub> accounts for less than 20 percent of the photochemical O<sub>3</sub> loss in the low stratosphere at midlatitudes, NO and NO<sub>2</sub> are vital in regulating the abundance of hydrogen and halogen radicals and thus the total photochemical O<sub>3</sub> destruction rate (WMO, 1994).

It should also be noted that there is observational evidence that tropospheric  $O_3$  (about 10 percent of the total-column  $O_3$ ) has increased in the Northern Hemisphere (north of 20 degrees N) over the past 3 decades (WMO, 1994). The upward trends are highly regional. Tropospheric  $O_3$  (and aerosols) can decrease global UV-B irradiances. However, recent trends in tropospheric  $O_3$  decreases (WMO, 1994).

#### $N_2O$

The major sources of  $N_2O$  emissions are described in the "Global Warming" section of this document. As noted previously,  $N_2O$  has an atmospheric lifetime of 150 years. The major sink of  $N_2O$  is photo dissociation following diffusion into the stratosphere (WMO, 1994). Its products of dissociation are the major source of stratospheric  $NO_x$ , which are important in regulating stratospheric  $O_3$ . The dissociation produces NO, which leads to the subsequent chemical destruction of stratospheric  $O_3$  (Kramlich and Linak, 1994). Formation of NO in the stratosphere is the result of photolysis of  $N_2O$  and reaction with an oxygen atom. The NO formed

subsequently reacts with stratospheric  $O_3$ , forming oxygen ( $O_2$ ). The net effect is that increased global concentrations of  $N_2O$  contribute to the thinning of the stratospheric  $O_3$  layer (Vitousek, 1997).

 $N_2O$  is emitted by natural and anthropogenic sources, which are listed in Table III-2 and have large uncertainties. Stratospheric  $O_3$  destruction is further complicated by potential interaction between chlorine monoxide and  $NO_2$ , which may decrease stratospheric  $O_3$  destruction by interfering with atomic chlorine formation (Kramlich and Linak, 1994). Since the mid-1970s, systematic tropospheric measurements of  $N_2O$  have been made at locations worldwide. These data show the atmospheric concentration of  $N_2O$  to be increasing at an average rate of approximately 0.3 percent per year (Khalil and Rasmussen, 1992; Vitousek et al, 1997)).

# $NO_r$

Emissions from aircraft are a relatively small source of  $NO_x$ . The impact of the aircraft emissions depend on the altitude as well as the amount of the emissions. Models indicate that the  $NO_x$  emissions from the current subsonic fleet produce upper-tropospheric  $O_3$  increases as much as several percent, maximizing at northern midlatitudes. In contrast, projected fleets of supersonic aircraft may decrease total-column  $O_3$  concentrations by 0.3-1.8 percent for the Northern Hemisphere (WMO, 1994).

# **Emissions Control Programs**

As noted in the previous section concerning global warming, the development of an emissions control program to achieve a specific environmental goal may need to take into account the impact on both  $NO_x$  and  $N_2O$  emissions and the resultant environmental impacts that may go beyond the specific goal. Some  $NO_x$  control programs strive to prevent  $NO_x$  emissions in the first place. Other control programs are directed at  $N_2O$  emissions, such as in adipic acid production. However, many  $NO_x$  control procedures for mobile and stationary sources convert NO into  $N_2$  and, in that process, a portion of the reactions will form  $N_2O$ .

#### References

Kramlich, J.C. and Linak, W.L, "Nitrous Oxide Behavior in the Atmosphere, and in Combustion and Industrial Systems," *Prog. Energy Combust. Sci.*, Vol 20, 1994.

Levine, J.S., "The Global Atmospheric Budget of Nitrous Oxide, and Supplement: Global Change: Atmospheric and Climatic," 5th International Workshop on Nitrous Oxide Emissions, Tsukuba, Japan, 1992.

Khalil, M.A. and Rasmussen, R.A., "The Global Sources of Nitrous Oxide," J. Geophys. Res. 97, 14651-14660, 1992.

Tie, X.X., G.P. Brasseur, B. Briegleb, and C. Grainier, "Two-Dimensional Simulation of Pinatubo Aerosol and Its Effect on Stratospheric Ozone," *J. Geophys. Res.*, 99, 20545-20562, 1994.

Vitousek, P.M., J. Aber, R.W. Howarth, G.E. Likens, P.A. Matson, D.W. Schindler, W.H. Schlesinger, and G.D. Tilman, "Human Alteration of the Global Nitrogen Cycle: Causes and Consequences," *Issues in Ecology*, Number 1, Spring 1997.

U.S. Environmental Protection Agency, Office of Policy, Planning and Evaluation, *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-1994*, November 1995, EPA-230-R-96-006.

World Meteorological Organization, *Scientific Assessment of Ozone Depletion: 1994*, Global Ozone Research and Monitoring Project--Report No. 37.

# E. Terrestrial Ecosystems

# N Deposition

Over the last 40 years, the rate atmospheric deposition of N compounds has increased more than 10 times in eastern North America (Vitousek, 1994). N accumulates in watersheds with high N deposition. The primary sources of N ( $NO_x$  and ammonia,  $NH_3$ ) deposition are the combustion of fossil fuels, the manufacture and use of fertilizers, livestock, and burning of biomass as a result of changing land use practices (Townsend, et al, 1996; Schlesinger and Hartley, 1992). Because North American terrestrial ecosystems are generally considered to be Nlimited, N deposition often has a fertilizing effect, accelerating plant growth. While this effect is often considered beneficial, N deposition is causing important adverse changes in some terrestrial ecosystems, including shifts in plant species composition and decreases in species diversity or undesirable nitrate leaching to surface and ground water and decreased plant growth.

# The N Cycle

All organisms require N to live; it is an essential component of cholorphyll, genetic material, and proteins. In many ecosystems on land and sea, the supply of N is a key factor controlling the nature and diversity of plant life, the population dynamics of both grazing animals and their predators, and vital ecological processes such as plant productivity and the cycling of carbon and soil minerals (Vitousek et al, 1997).

Although the earth's atmosphere is 78 percent N gas (in the form of the  $N_2$  molecule), most plants and animals cannot use the atmospheric  $N_2$  directly. Before plants can use N, it must be bonded ("fixed") into inorganic compounds, mainly ammonium,  $NH_4^+$ , and nitrate,  $NO_3^-$ . There are both natural and anthropogenic processes that "fix" atmospheric  $N_2$  into these inorganic compounds.

The main natural source of N fixation is N fixing organisms. N fixing organisms include algae and bacteria. The most important ones are bacteria that form symbiotic relationships with higher plants, especially legumes. These bacteria manufacture an enzyme that enables them to convert atmospheric  $N_2$  directly into plant-usable forms. Lightning is also a natural source of N fixation as conditions of high pressure and temperature allow  $N_2$  and  $O_2$  to combine and form into nitrates.

During the past century, human activities have at least doubled the rate of transfer of atmospheric  $N_2$  to biologically usable forms (Vitousek et al, 1997; Schlesinger, 1992). The major anthropogenic sources include industrial processes that produce N fertilizers, the combustion of fossil fuels, and the cultivation of legumes. Furthermore, biomass burning, drainage of wetlands and land clearing are important activities that contribute to biologically available N. Most of this

N is deposited by precipitation over land, where it enters biogeochemical cycles (Schlesinger, 1992).

# N Deposition Effects on Forest Ecosystems

Rates of growth in North American forest ecosystems have traditionally been considered limited by N availability and, thus, N inputs are usually considered beneficial. In N-limited forests, most N is bound in soil organic matter and only becomes available for biotic uptake when decomposition of soil organic matter releases inorganic nitrate and ammonium ions. Plants and microbes promptly absorb inorganic N, maintaining minimal available N pools in the soil and thereby limiting leaching losses. Although N limitation is still widespread, recent findings in North America and Europe suggest that, because of chronic N deposition from air pollution, some forests are no longer N-limited and that this condition may increase tree mortality and alter water quality (Aber et al, 1996; Sullivan, 1993; Vitousek et al, 1997).

N saturation has a complex cascade of damaging effects for ecosystems (Vitousek et al, 1997). As  $NH_4^+$  builds up in the soil, it is increasingly converted to nitrate, a process that releases hydrogen ions and helps acidify the soil. The buildup of nitrate may result in leaching of nitrate into streams or groundwater. As negatively charged nitrates leach away, they carry with them positively charged alkaline minerals, thus decreasing soil fertility. As calcium is depleted and the soil acidified, aluminum ions are mobilized, eventually reaching toxic concentrations that can damage tree roots or kill fish if the aluminum washes into streams.

The impacts of N saturation first became apparent in Europe almost two decades ago when scientists observed significant increases in nitrate concentrations in some lakes and streams and also extensive yellowing and loss of needles in spruce and other conifer forests subjected to heavy N deposition (Vitousek et al, 1997). In Southern California, elevated nitrate and NO fluxes from soil, indicators of N saturation, have recently been reported for chaparral watersheds in the San Gabriel Mountains as well as in portions of the mixed conifer forest in the San Bernardino Mountains (Fenn et al, 1996). While there are important differences in the magnitude of N deposition, vegetation and soil cover, areas of concern include the Great Smoky Mountains in North Carolina, northeastern United States, southern California, and the Colorado Front Range of the Rocky Mountains (Johnson et al, 1991; van Miegroet et al, 1993; Fenn et al, 1996; Williams et al, 1996; Vitousek et al, 1997). In some cases excess N availability can lead to decreased tree growth and increased mortality (McNulty et al.1996). Nutrient imbalances, with signs such as depressed Ca:Al and Mg:N ratios in foliage, may act with other stresses to produce these effects (Aber et al. 1996).

N saturation has been defined in several ways, including, as a condition in which (1) available N is frequently in excess of total biotic demand, (2) vegetation within an ecosystem no longer exhibits a positive growth response to N addition, even through other growth factors are not growth limiting, and (3) sustained N losses approximate or exceed N inputs--the N retention capacity of the system has been exceeded (Fenn, et al, 1996). Recent research suggests that because N-limited forests retain most N inputs, N deposition above relatively low amounts to

these systems is largely cumulative (Ryan, 1996). Moreover forests respond nonlinearly to cumulative N inputs if elevated N deposition continues over long periods. That is, N inputs to N-limited forests (with little accumulated N) stimulate their growth and those inputs are strongly retained with little nitrate leaching. After N inputs have accumulated (to cause N saturation), the response of the forest changes so that N deposition increases nitrate leaching and may decrease forest growth. In order to predict how an ecosystem will respond to N deposition one needs to know where the ecosystem is on this response curve to cumulative N inputs. Predictions are uncertain about how much cumulative N inputs it will take over what time period to bring about N saturation in particular forest ecosystems because processes controlling N accumulation and loss are not completely understood and because data on the current stage of N saturation are lacking for most forests.

# N Deposition Effects on Grassland Ecosystems Reduced Species Diversity

Most natural ecosystems have been N limited. Accordingly, native plant species have adapted to this environmental constraint. With an increase in N deposition, many of the native plant species may no longer be able to compete with other species that are adapted to high N conditions.

N deposition in grassland ecosystems has been shown to (1) alter the composition of the grassland species in the affected area, (2) decrease species diversity, and (3) increase the above ground productivity (Wedin and Tilman, 1996). In a 12 year experimental study of N deposition on Minnesota grasslands, plots dominated by native grasses shifted to low-diversity mixtures at all but the lowest N addition rates (Wedin and Tilman, 1996). While prairie grasses can thrive were N availability is limited, as N availability is increased, competing, non-native species begin invading the prairie plots. After 12 years of N addition, species richness (number of plant species per area) declined by more than 50 percent, with the greatest losses at levels spanning current atmospheric deposition rates in eastern North America (Wedin and Tilman, 1996). That is, the native grasses with supplemental N deposition showed an impaired ability to compete with non-native species. In England, N fertilizers applied to experimental grasslands led to increased dominance by a few N responsive grasses and loss of many other plant species; at the highest fertilization rate, the number of plant species declined more than fivefold (Vitousek et al, 1997).

# **Global Warming Impacts**

Because plants use atmospheric carbon dioxide to fix carbon in their tissues, over their lifetime they are a sink for atmospheric carbon dioxide, the primary global warming gas. As the plants die and decompose, a portion of the carbon returns to the atmosphere and a portion may be incorporated into the soils, resulting in a net sink of carbon dioxide. With increased N inputs and resulting increased plant growth, it has been hypothesized that the amount of carbon removed from the atmosphere might increase.

There is considerable uncertainty over both the magnitude and persistence of any Nderived carbon sink. Different ecosystem types vary greatly in their potential for carbon storage. Much of the area experiencing high N deposition is covered by grasslands or cultivated areas. In general, N deposition that stimulates wood production will cause a relatively large and long-term removal of carbon from the atmosphere. In contrast, foliar biomasses in forests and grasslands have much more limited capacity for carbon storage and deposition onto cultivated areas is not likely to contribute to any terrestrial sink (Townsend, et al, 1996).

In the case of the native prairie grasses, the decomposition is slow, resulting in the storage of carbon in the earth. In contrast, the non-native plants which thrive on N (and replace native plants under increased N deposition), decompose rapidly due to their high N content and, thus, return most of their carbon to the atmosphere, where it can contribute to global warming. The 12 year study indicates that the carbon storage, in some plots, was decreased by 50 percent and concludes that N-caused shifts in species composition limit the ability of temperate grasslands to serve as significant long-term carbon stores (Wedin and Tilman, 1996). Thus, N deposition shifted the mix of plants toward the faster growing non-native species which decreased the area's ability to move carbon dioxide from the air and store it in the soil.

References:

Aber, John D., Knute J. Nadelhoffer, Paul Steudler, and Jerry M. Melillo. 1989. Nitrogen Saturation in Northern Forest Ecosystems. *BioScience* Vol. 39 No. 6. June, 378-386.

Aber JD, Melillo JM, Nadelhoffer KJ, Pastor J, Boone RD. 1991. "Factors controlling nitrogen cycling and nitrogen saturation in northern temperate forest ecosystems." *Ecol. Appl.* 1:305-315

Aber, J.D., A. McGill, S. G. McNulty, R.D. Boone, K.J. Nadelhoffer, M. Downs, R. Hallett. "Forest biogeochemistry and primary production altered by nitrogen saturation." *Water, Air, & Soil Pollution*\_volume 85 (p.1665-1670),1995.

Agren GI, Bosatta E. 1988. Nitrogen saturation of terrestrial ecosystems *.Environ. Pollut.* 54:185-97.

Christ M, Zhang Y, Likens GE, Driscoll CT. 1995. Nitrogen retention capacity of a northern hardwood forest soil under ammonium sulfate additions. *Ecol. Appl.* 5:802-12.

Fenn, M., M.A. Poth, D.W. Johnson. "Evidence for nitrogen saturation in the San Bernardino Mountains in Southern California," *Forest Ecology and Management* 82 (p. 211-230), 1996.

McNulty, S.G., J.D. Aber, S.D. Newman. 1996. Nitrogen saturation in a high elevation New England spruce-fir stand. *Forest Ecology and Management* 84:109-121.

Ryan, Douglas F., J.D. Aber, J.S. Baron, B.T. Bormann, C.T. Driscoll, M.E. Fenn, D.W. Johnson, A.D. Lemly, S.G. McNulty, and R. Stottlemyer, "Nitrogen Deposition from Air Pollution: Implications for Forest Ecosystems in North America" draft 1996.

Schlesinger, W.H., Biogeochemistry: An Analysis of Global Change, 1991.

Schlesinger, W.H. and Hartley, A.E., "A Global Budget for Atmospheric NH<sub>3</sub>", *Biogeochemistry* 15: 191-211, 1992.

Stoddard JL. 1994. Long-term changes in watershed retention of nitrogen: Its causes and aquatic consequences. In: *Environmental Chemistry of Lakes and Reservoirs, Advances in Chemistry Series No. 237*, ed. LA Baker, pp. 223-284, Washington, DC: Amer. Chem. Soc.

Sullivan, T.J. 1993. "Whole ecosystem nitrogen effects research in Europe." *Environmental Science & Technology* 37:1482-1486.

Townsend, A.R., B.H.Braswell, E.A.Holland, and J.E.Penner, "Spatial and Temporal Patterns in Terrestrial Carbon Storage Due to Deposition of Fossil Fuel Nitrogen," *Ecological Applications*, 6(3), 1996 pp. 806-814, 1996.

Van Sickle, J. and M.R. Church. 1995. *Methods for estimating the relative effects of sulfur and nitrogen deposition on surface water chemistry*. EPA Rep. no. EPA/600/R-95/172. Washington, DC US Environ. Protect. Agency.121 pp.

Vitousek, P.M., J. Aber, R.W. Howarth, G.E. Likens, P.A. Matson, D.W. Schindler, W.H. Schlesinger, and G.D. Tilman, "Human Alteration of the Global Nitrogen Cycle: Causes and Consequences," *Issues in Ecology*, Number 1, Spring 1997.

Wedin, David A. and David Tilman, "Influence of Nitrogen Loading and Species Composition on the Carbon Balance of Grasslands." *Science*, Volume 274, December 6, 1996.

Williams, Mark W., Jill S. Baron, Nel Caine, Richard Sommerfield and Robert Sanford, Jr. 1996. "Nitrogen Saturation in the Rocky Mountains." *Environmental Science & Technology* Vol 30 No 2.

Vitousek, P.M., *Ecology*, Volume 75 (1861),1994.

#### F. Toxic Products

#### Introduction

In the atmosphere,  $NO_x$  reacts with common hydrocarbons and  $O_3$  to form a wide variety of toxic compounds. Shepson et al. (1987) reported that photochemical reactions in a laboratory smog chamber substantially increased the mutagenicity of such mixtures. This added mutagenicity, measured by Ames assays, was time-dependent and associated mostly with the gas phase. The mutagenic transformation products were also refractory, accumulating and persisting in the chamber for hours after their production.

The accumulation of new mutagens by photochemically-mediated nitrate reactions raises significant public health concerns. Chemicals that are able to alter bacterial genes (DNA) also have the potential to cause mutations in higher organisms, and some fraction of these mutations may initiate carcinogenesis. Examples of transformation products thought to contribute to increased mutagenicity include the nitrate radical, peroxyacetyl nitrate, nitroarenes, and nitrosamines (Shepson et al., 1987; CARB, 1986). Toxicological data on many of these compounds are limited, making it difficult to discuss effects of specific reaction products, but the overall trend is well-established. Information on formation and toxicity of groups of substances is summarized below.

#### Nitrate Radical

The gaseous nitrate radical is a product of the reaction of  $NO_2$  with  $O_3$  (Finlayson-Pitts and Pitts, 1993). This compound dissociates rapidly in sunlight, but it is possible that nighttime concentrations may become significant. Although health effects of the gaseous nitrate radical have not been described, radicals are generally highly reactive compounds with the potential to damage complex biological molecules such as proteins, lipids, and nucleic acids. Pitts et al. (1983) have postulated that the nitrate radical is capable of inducing genetic changes, but this does not appear to have been tested in the laboratory.

# Peroxyacetyl Nitrate (PAN)

PAN (CH<sub>3</sub>CO<sub>3</sub>NO<sub>2</sub>) is an organic-nitrogenous air pollutant formed by complex photochemical reactions of common aliphatic compounds and NO<sub>x</sub>. PAN has a relatively long thermal decomposition lifetime in the absence of NO. When NO is removed from the atmosphere by reaction with O<sub>3</sub>, as in the afternoon and above the mixed layer at night, PAN can persist. PAN is an important lacrimator, thought to contribute much of the eye-stinging effect of urban smog. Tests for mutagenicity and carcinogenicity of pure PAN have not been reported in the literature. However, Shepson et al. (1987) concluded that PAN formation accounted for a substantial part of the increased mutagenicity observed in their smog chamber study.

#### Nitroarenes

Butler et al. (1981) demonstrated that polynuclear aromatic hydrocarbons (PAHs) are nitrated in the laboratory to form nitroarenes when exposed to concentrations of  $NO_2$  and nitric acid similar to those found in ambient air. These results suggest that PAHs (hydrocarbons having multiple carbon-ring structures, and major constituents of polycyclic organic matter) may also be rapidly nitrated in the atmosphere. Rosenkrantz and Mermelstein (1983) reported similar reactions in PAHs adsorbed on soot particles and other substrates. Many of these compounds are mutagens and/or carcinogens, even when their parent non-nitrated PAH analogs are not. Examples from this broad spectrum of substances include nitrated analogs of quinoline, pyrene, fluorine, and naphthalene. Rosenkrantz and Mermelstein (1983) reported that many nitroarenes are potent bacterial mutagens and also produce a variety of genetic and genotoxic effects in mammalian cell assays, including unscheduled DNA synthesis, sister-chromatid exchange, chromosomal aberrations, gene mutations, and cell transformation. Data from whole animals are limited, but various nitroarenes have produced skin tumors in mice and bladder tumors in dogs and monkeys.

# Nitrosamines

Gehlert et al. (1979) reported the formation of nitrosamines (nitrated organic amine compounds) from amines and  $NO_x$  in the laboratory, and proposed that this reaction also occurs in the atmosphere. Although nitrosamines are rapidly decomposed by sunlight, ambient concentrations could rise during the night. Nitrosamines are a major class of powerful chemical carcinogens, with different compounds sometimes exhibiting high target organ specificity in animal studies. Sites of cancers induced by nitrosamines in rodents include the liver, bladder, lung, kidney, and pancreas (Casarett and Doull, 1986). Carcinogenesis occurs through metabolic activation, followed by methylation of DNA by the electrophilic metabolites.

#### Summary

In laboratory tests, nitrates react readily with common organic chemicals, and even  $O_3$ , in the presence of light to form a wide variety of mutagenic and carcinogenic transformation products. Although animal inhalation studies of individual compounds formed in this way are limited, results of bacterial and mammalian cell bioassays indicate clearly that both mixtures and individual mixture components are able to alter DNA. More research, especially whole animal studies, would help the EPA understand the potential magnitude of public health impacts of these transformation products.

#### References

Butler, J.D. and P. Crossley. Atmospheric Environment. 15:91 (1981).

California Air Resources Board. *The effects of oxides of nitrogen on California air quality*. March 1986.

Casarett, I. and L.J. Doull. *Toxicology*. Macmillan Publishing Co., New York. C.D. Klaassen, M.O. Amdur, and J. Doull (eds.) (1986).

Finlayson-Pitts, B.J. and J.N. Pitts, Jr., "Atmospheric Chemistry of Tropospheric Ozone Formation: Scientific and Regulatory Implications," *Air and Waste Management Association*, Vol. 43, August 1993.

Gehlert and W. Rolle. Experimentia 33:579 (1979).

Pitts, Jr. J.N., A.M. Winer, G.W. Harris, W.P.L. Carter, and E.C. Tuazon. Trace nitrogenous species in urban atmospheres. *Environ. Health Perspectives* 52:153-157 (1983).

Rosencrantz, H.S. and R. Mermelstein. Mutagenicity and genotoxicity of nitroarenes: all nitrogen-containing chemicals were not created equal. *Mutation Research* 114: 217-267 (1983).

Shepson, P.B., T.E. Kleindienst, and E.O. Edney. *The production of mutagenic compounds as a result of urban photochemistry*. EPA 600/3-87/020 (1987).

#### IV. Interprogram Issues

# A. Local and Regional NO<sub>x</sub> Requirements

#### Policy Decisions Should Consider All the Effects of NO<sub>x</sub> Emissions

As described in detail in sections II and III of this document,  $NO_x$  emissions impact the environment in a number of ways. Some of these are regional scale, adverse impacts. As noted in the "Ozone" section, decreasing local  $NO_x$  emissions can, in some cases, increase local  $O_3$ concentrations. This effect of  $NO_x$  emissions reductions must be carefully weighed against the multiple beneficial effects of  $NO_x$  emissions reductions with respect to acid deposition, drinking water nitrate, eutrophication, global warming,  $NO_2$ , N in forest ecosystems,  $O_3$ , PM, stratospheric  $O_3$  depletion, toxics and visibility degradation. Further, the effects of emissions of  $N_2O$  are also of concern with respect to global warming and stratospheric  $O_3$  depletion. The EPA believes that policy decisions should be made considering the environment as a whole, rather than narrowly viewing each program, one at a time. As stated by two committees of the National Research Council:

Changes in VOCs and  $NO_x$  will, because of their complex chemical interactions, also lead to changes in a variety of other pollutants associated with  $O_3$ , such as nitric acid, peroxyacetyl nitrate,  $NO_2$ , and aerosol particles. Some of these pollutants have known harmful effects on human health and welfare. Hence, it is important to recognize that control strategies implemented for  $O_3$  will simultaneously affect other species. (NRC, 1991).

Visibility is just one of many air-quality problems. The pollutants that impair visibility contribute to other environmental problems, some of which have been or are being considered as objects of federal, state, or local legislation or regulation. For example, controls aimed at decreasing acid deposition or lowering ambient concentrations of  $O_3$  and  $PM_{10}$  could improve visibility in Class I areas; conversely, controls aimed at improving visibility could alleviate other air-quality problems. Policy makers should weigh these linkages in the design and assessment of possible control strategies. (NRC, 1993).

The remainder of this section primarily describes (1) the effects of NO<sub>x</sub> emissions reductions on O<sub>3</sub> concentrations, (2) CAA requirements for NO<sub>x</sub> controls in certain areas, (3) EPA action to waive these requirements in certain cases, and (4) the need for regional scale NO<sub>x</sub> controls in the future for purposes of O<sub>3</sub> attainment. Although the focus of the discussion is on O<sub>3</sub>, EPA intends to use its discretion wherever possible to assure that policy decisions concerning O<sub>3</sub> are consistent with other environmental goals which may suggest the need for reductions in NO<sub>x</sub> emissions. The EPA believes that effective O<sub>3</sub> control requires an integrated strategy that combines cost-effective reductions in emissions at the local, state, regional, and national levels.

# Role of $NO_x$ and VOCs in Urban $O_3$

Great progress has been made over the past two decades, at the local, state and national levels in controlling emissions from many sources of air pollution. During this period, control of VOCs was the main strategy employed in efforts to lower ground-level  $O_3$ . The 1970 and 1977 amendments to the CAA did not explicitly require NO<sub>x</sub> emissions reductions from stationary sources for purposes of attainment of the  $O_3$  standard. With respect to mobile sources, both VOC and NO<sub>x</sub> emissions were decreased substantially. Scientific evidence at the time suggested that VOC emissions reductions were preferred in most instances (EPA, 1993). The VOC control approach was reinforced by the fact that NO<sub>x</sub> emissions decreases could in some cases increase  $O_3$  concentrations.

More recently, it has become clearer that  $NO_x$  controls may be needed in many areas, especially areas where  $O_3$  concentrations continue to be high over a large region (as in the Midwest and Northeast). In the debate leading up to the 1990 CAA amendments, Congress included consideration of  $NO_x$ . A report by the Office of Technology Assessment (U.S. Congress, 1989) provided support for inclusion of  $NO_x$  controls in the  $O_3$  program. The 1990 amendments changed the statutory framework to place  $NO_x$  emissions reductions on a more equal footing with the VOC emissions reductions.

In the process of adding these new NO<sub>x</sub> requirements, Congress recognized that NO<sub>x</sub> emissions reductions would help achieve lower O<sub>3</sub> concentrations in some O<sub>3</sub> nonattainment areas, but that "there are some instances in which NO<sub>x</sub> reductions can be of little benefit in reducing O<sub>3</sub> or can be counter-productive, due to the offsetting ability of NO<sub>x</sub> to `scavenge' (i.e., react with) O<sub>3</sub> after it forms" (H.R. Rep. No. 490, 101st Congress, 2nd Sess., at 204). The Congress provided for additional review and study under section 185B of the CAA "to serve as the basis for the various findings contemplated in the NO<sub>x</sub> provisions" (H.R. Rep. 490 at 257).

Under section 185 of the CAA, the EPA, in conjunction with the National Academy of Sciences, conducted a study on the role of  $O_3$  precursors in tropospheric  $O_3$  formation which examined the role of  $NO_x$  and VOC emissions, the extent to which  $NO_x$  emissions reductions may contribute or be counterproductive to achieving attainment in different nonattainment areas, the sensitivity of  $O_3$  to the control of  $NO_x$ , the availability and extent of controls for  $NO_x$ , the role of biogenic VOC emissions, and the basic information required for air quality models. The NAS portion of the study was published in 1991 (NRC, 1991). The section 185B study was completed and submitted to Congress July 30, 1993 (EPA, 1993).

The 1991 National Academy of Sciences report recommends that "To substantially reduce  $O_3$  concentrations in many urban, suburban, and rural areas of the United States, the control of  $NO_x$  emissions will probably be necessary in addition to, or instead of , the control of VOCs." The section 185B study concludes that the  $O_3$  precursor control effort should focus on  $NO_x$  controls in many areas and that the analysis of  $NO_x$  benefits is best conducted through photochemical grid modeling. The shift to consideration of both  $NO_x$  and VOCs, explains the

section 185B study, coincides with improved data bases and modeling techniques that provide the analytical means to evaluate the effectiveness of  $O_3$  precursor control strategies.

# The 1990 Clean Air Act Amendments (CAAA)

The 1990 CAAA include substantial new requirements to decrease emissions of NO<sub>x</sub> from major stationary sources. To help attain the 1-hour O<sub>3</sub> air quality standard in the near-term, section 182(f) of the CAAA requires certain existing sources to install reasonably available control technology (RACT) and new sources must install controls representing the lowest achievable emission rate (EPA, 1992). Section 182(f) also specifies circumstances under which the new NO<sub>x</sub> requirements for RACT and NSR would be limited or would not apply. To decrease acid deposition, the CAAA require coal-fired utility boilers to meet emission limits in two phases. Further, in a longer-term provision, the CAAA require States to adopt additional control measures as needed to attain the O<sub>3</sub> standard. This requirement supplements the RACT and NSR requirements. Thus, a State would need to require NO<sub>x</sub> controls which achieve emissions reductions greater than the NO<sub>x</sub> RACT/phase I acid deposition limits where additional reductions in emissions of NO<sub>x</sub> are necessary to attain the O<sub>3</sub> standard by the attainment deadline.

For  $O_3$  nonattainment areas designated as serious, severe, or extreme, state attainment demonstrations involve the use of photochemical grid modeling for each nonattainment area. Although these attainment demonstrations were due November 15, 1994, the magnitude of this modeling task, especially for areas that are significantly affected by transport of  $O_3$  and  $O_3$  precursors (NO<sub>x</sub> and VOCs) generated outside of the nonattainment area, has delayed many states in submitting complete modeling results. Recognizing these challenges, EPA issued guidance on  $O_3$  demonstrations, based on a two-phase approach for the submittal of  $O_3$  SIP attainment demonstrations (Nichols, 1995). The guidance established a 2-phase approach which includes an intensive modeling effort to address the problem of long distance transport of  $O_3$ , NO<sub>x</sub> and VOCs and submittal of the attainment plans in 1997.

# Section 182(f) of the CAA-- $NO_x$ Waiver

As described in EPA guidance (EPA, December 1993), the CAAA include new provisions in section 182(f) to control emissions of  $NO_x$  and specify circumstances under which the new  $NO_x$  requirements would be limited or would not apply. Section 182(f)(1) provides that certain new  $NO_x$  requirements shall not apply if the Administrator determines that any one of the following tests is met:

- (1) in any area, the net air quality benefits are greater in the absence of NO<sub>x</sub> reductions from the sources concerned;
- (2) in nonattainment areas not within an  $O_3$  transport region, additional  $NO_x$  reductions would not contribute to  $O_3$  attainment in the area; or
- (3) in nonattainment areas within an  $O_3$  transport region, additional  $NO_x$  reductions would not produce net  $O_3$  air quality benefits in the transport region.

Further, section 182(f)(2) states that the application of the new NO<sub>x</sub> requirements may be limited to the extent necessary to avoid excess reductions of NO<sub>x</sub> as determined by applying tests similar to tests (1)-(3) above.

#### Administrative Procedures

Section 182(f)(3) provides that a person may petition the Administrator for a  $NO_x$  exemption at any time after the final section 185B report is submitted to Congress. The final section 185B report was sent to Congress by the Administrator on July 30, 1993. The petition may be made with respect to any nonattainment area or any  $O_3$  transport region. The EPA must grant or deny a petition within 6 months after its filing.

If EPA grants a petition, the section  $182(f)NO_x$  requirements or portions of those requirements (EPA, 1992), would no longer apply to those sources or areas, as described in EPA's approval action. However, States remain free to adopt  $NO_x$  restrictions for other reasons. For example, a State may determine that  $NO_x$  emissions reductions are needed for purposes of  $O_3$ maintenance planning,  $O_3$  attainment in separate downwind nonattainment areas, visibility protection, PM control strategy, acid deposition program or other environmental protection.

The CAA requires EPA to view  $NO_x$  waivers in a narrow manner. In general, section 182(f) provides that waivers must be granted if states show that decreasing  $NO_x$  within a nonattainment area would not contribute to attainment of the  $O_3$  NAAQS within the same nonattainment area (Seitz, 1995). Only the role of local  $NO_x$  emissions on *local attainment* of the  $O_3$  standard is considered in nonattainment areas outside an  $O_3$  transport region. The role of  $NO_x$  in *regional attainment* will be addressed separately. As described in the "Ozone" section,  $NO_x$  has been shown to be effective in decreasing regionally transported  $O_3$ .

#### Status of NO<sub>x</sub> Waiver Petitions

In response to State  $NO_x$  waiver petitions submitted between 1992-1995, EPA granted  $NO_x$  waivers under section 182. Most waivers were granted on the basis that the area had already attained the  $O_3$  standard and, thus, additional  $NO_x$  (or VOC) reductions "would not contribute to ozone attainment in the area." In some cases, the waivers were granted based on dispersion modeling which showed that the area would attain just as expeditiously based solely on additional VOC reductions or that local  $NO_x$  reductions increased local peak  $O_3$  concentrations; this also meets the above test that additional  $NO_x$  reductions would not contribute to  $O_3$  attainment in the area.

Specifically, the EPA received petitions for a  $NO_x$  waiver for 51  $O_3$  nonattainment areas. Of these petitions, EPA approved (as of July 1997) waivers for 48 nonattainment areas and 3 were pending. Most of the waivers granted (28) were simply based on air quality monitoring data over a period of 3 or more years indicating the area had attained the  $O_3$  standard (and, thus, additional  $NO_x$  reductions were not needed for attainment). Several States submitted  $NO_x$  waiver petitions (7) accompanied by an attainment plan showing achievement of the  $O_3$  standard by the statutory deadline through additional VOC controls only. None of these 41 nonattainment areas with approved NO<sub>x</sub> waivers have demonstrated or even sought to demonstrate that NO<sub>x</sub> reductions might increase O<sub>3</sub> concentrations in specific areas. Only in the cases of the Lake Michigan (9 nonattainment areas), Phoenix AZ, Baton Rouge LA and the Houston/Beaumont TX areas was information submitted to show that, on some days, local NO<sub>x</sub> emissions decreases lead to local increases in peak O<sub>3</sub> concentrations in some but not necessarily all portions of these areas. Even for the 4 areas with modeling information, those analyses were generally considered preliminary analyses that would be replaced with more complete modeling associated with attainment plans.

# NO<sub>x</sub> waivers are granted on a temporary or contingent basis

The EPA's approval of any  $NO_x$  exemption is granted on a contingent basis (Seitz, 1994). That is, a monitoring-based exemption lasts for only as long as the area's monitoring data continue to demonstrate attainment. Thus, if a violation is monitored (prior to the area being redesignated as being in attainment) the exemption would need to be revoked and the requirement to adopt  $NO_x$  controls would again apply. Similarly, any modeling-based exemption may need to be withdrawn if updated modeling analyses reach a different conclusion than the modeling on which the exemption was based (EPA, 1992).

In the <u>Federal Register</u> notices approving individual waiver petitions, EPA gave notice that approval of the local petition is on a contingent or temporary basis and stated that additional local and regional NO<sub>x</sub> emissions reductions may be needed to reduce the long range transport of O<sub>3</sub>. Where such additional NO<sub>x</sub> reductions are necessary to reduce the long range transport of O<sub>3</sub>, EPA stated that authority provided under section 110(a)(2)(D) of the CAA would be used and that a section 182(f) NO<sub>x</sub> waiver would, in effect, be superseded for those control requirements needed to meet the section 110(a)(2)(D) action. Further, EPA noted that States may require additional NO<sub>x</sub> reductions in these nonattainment areas for non-O<sub>3</sub> purposes, such as attainment of the PM-10 standard or achieving acid rain reduction goals.

In some cases, despite a potential increase in  $O_3$  concentrations in central urban areas, State and local agencies may need to decrease  $NO_x$  emissions as part of a larger plan to meet various environmental goals. For example, the South Coast area of California models this effect, yet substantial  $NO_x$  emissions reductions are contained in their attainment plan. The  $NO_x$ emissions reductions in the South Coast are needed to attain the  $PM_{10}$  standard and to maintain the  $NO_2$  standard in the same air basin as well as to help lower  $O_3$  concentrations in areas downwind of the basin. In a different situation,  $NO_x$  emissions reductions in the New York metropolitan area are needed for downwind areas within the State and in other States to attain the  $O_3$  standard; yet, additional VOC controls may be needed in the metropolitan area to offset the local impact of  $NO_x$  emissions reductions. Similarly,  $NO_x$  emissions reductions in areas upwind of the Northeast Ozone Transport Region may be needed to help downwind areas attain and maintain the  $O_3$  standard, even though those  $NO_x$  emissions reductions may not help the upwind areas lower local  $O_3$  concentrations. Models provide a way to test various control strategies so that the best approach, considering all the environmental goals, can be selected. Further, the NO<sub>x</sub> waiver does not shield an area from the acid deposition requirements of Title IV of the CAA. Regional and/or local NO<sub>x</sub> emissions reductions may also be needed to slow eutrophication in sensitive water bodies, improve visibility and/or decrease PM concentrations. Furthermore, increases in NO<sub>x</sub> are not viewed by EPA as a solution to local O<sub>3</sub> problems; NO<sub>x</sub> emissions reductions are generally needed at least to counter increases in NO<sub>x</sub> emissions due to economic growth. Thus, a local NO<sub>x</sub> waiver should be considered temporary and does not shield an area from NO<sub>x</sub> requirements needed for O<sub>3</sub> attainment in downwind areas or to meet other CAA requirements.

#### Modeling Analyses

The OTAG addressed the complex issue of regional impacts due to transport of  $NO_x$  and VOC emissions. The OTAG modeling results indicate that urban  $NO_x$  reductions produce widespread decreases in  $O_3$  concentrations on high  $O_3$  days. In addition, urban  $NO_x$  reductions also produce limited increases in  $O_3$  concentrations locally, but the magnitude, time, and location of these increases generally do not cause or contribute to high  $O_3$  concentrations. Most urban  $O_3$  increases modeled in OTAG occur in areas already below the  $O_3$  standard and, thus, in most cases, urban  $O_3$  increases resulting from  $NO_x$  reductions do not cause exceedance of the  $O_3$  standard. There are a few days in a few urban areas where  $NO_x$  reductions are predicted to produce  $O_3$  increases in portions of an urban area with high  $O_3$  concentrations.

In other words, modeling analyses conducted as part of the OTAG process indicated that, in general,  $NO_x$  reduction disbenefits are inversely related to  $O_3$  concentration. On the low  $O_3$ days leading up to an  $O_3$  episode (and sometimes the last day or so) the increases are greatest, and on the high  $O_3$  days, the increases are least (or nonexistent); the  $O_3$  increases occur on days when  $O_3$  is low and the  $O_3$  decreases occur on days when  $O_3$  is high. This indicates that, in most cases, urban  $O_3$  increases may not produce detrimental effects. However, OTAG modeling indicates that at least one area for one day of one episode experienced an increase in  $O_3$  on a high  $O_3$  day. Overall, OTAG modeling thus demonstrates that the  $O_3$  reduction benefits of  $NO_x$  control far outweigh the disbenefits of urban  $O_3$  increases in both magnitude of  $O_3$  reduction and geographic scope.

It should also be noted that the modeling analyses completed within the OTAG process necessarily utilized a larger grid size than States are likely to use in their attainment plans. That is, future analyses by States will likely use smaller grid sizes. The smaller grid sizes should provide more precise information on effects such as local NO<sub>x</sub> emissions reacting with local O<sub>3</sub>. Furthermore, new work is on-going to analyze air quality monitoring data, in part, to assess weekday and weekend patterns that may relate changes in NO<sub>x</sub> and VOC emissions to changes in O<sub>3</sub> concentrations. These air quality modeling and monitoring studies will provide additional information that may be important as States develop their attainment plans.

# Regional Transport of $O_3$

The problem of regional transport of  $O_3$  and its precursors is widely recognized by the States. In response to concerns about this problem raised by state environmental commissioners

comprising the Environmental Council of the States, EPA has worked closely with states in the OTAG to develop various recommended control measures intended to address the regional nature of  $O_3$ . Similarly, State and local air administrators passed a unanimous resolution endorsing national NO<sub>x</sub> emission regulations (Becker, 1995).

Control strategies need to consider efforts to decrease regional scale emissions as well as local emissions. In general,  $NO_x$  emissions decreases in upwind, rural areas coupled with VOC emissions reductions in urban nonattainment areas appears to be an effective strategy. In some cases however, the urban nonattainment area is also upwind of another urban nonattainment area or biogenic VOC emissions are very high. In these cases, local  $NO_x$  emissions reductions may be needed in addition to VOC emissions reductions for purposes of  $O_3$  attainment. In both cases, decreases in precursor emissions in the upwind areas will help the downwind metropolitan areas attain and maintain the  $O_3$  standard. Thus, effective  $O_3$  control will require an integrated strategy that combines cost-effective decreases in emissions at the local, state, regional, and national levels. Specific regional perspectives on the need for an integrated strategy are described in the "Ozone" section.

# References

Becker, William, Executive Director of STAPPA/ALAPCO, Letter to U.S. EPA Public Docket, October 13, 1995.

National Research Council, Committee on Haze in National Parks and Wilderness Areas, *Protecting Visibility in National Parks and Wilderness Areas*, 1993.

National Research Council, Committee on Tropospheric Ozone Formation and Measurement, *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, 1991.

Nichols, Mary D., Assistant Administrator for Air and Radiation, U.S. Environmental Protection Agency, "Ozone Attainment Demonstrations," memorandum to EPA Regional Administrators, March 2, 1995.

Ozone Transport Assessment Group, Joint meeting of RUSM & ISI workgroups, "First Round Strategy Modeling," October 25, 1996.

Seitz, John S., Director, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, "Section 182(f) Nitrogen Oxides (NO<sub>x</sub>) Exemptions—Revised Process and Criteria," EPA memorandum to Regional Air Directors, May 27, 1994.

Seitz, John S., Director, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, "Section 182(f) Nitrogen Oxides (NO<sub>x</sub>) Exemptions-- Revised Process and Criteria," EPA memorandum to Regional Air Directors, February 8, 1995.

US Congress, Office of Technology Assessment, Catching Our Breath: Next Steps for Reducing

Urban Ozone, Washington DC 1989.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *The Role of Ozone Precursors in Tropospheric Ozone Formation and Control--A Report to Congress*. EPA-454/R-93-024,1993.

U.S. Environmental Protection Agency, "Nitrogen Oxides Supplement to the General Preamble for Implementation of Title I of the Clean Air Act," <u>Federal Register</u> of November 25, 1992 (57 FR 55620).

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, "Guideline for Determining the Applicability of Nitrogen Oxides Requirements Under Section 182(f)," December 1993.

B. Timing of  $NO_x$  Emissions Reductions: Seasonal or Year-Round

# Regulation Can Affect the Seasonal Distribution of NO<sub>x</sub> Emissions

As noted previously,  $NO_x$  emissions have adverse impacts on the environment in several ways. In some cases, the timing of the  $NO_x$  emissions can be important to the subsequent environmental impacts. As noted below, year-round reductions in  $NO_x$  emissions are more helpful than seasonal approaches at minimizing the impacts of acid deposition and eutrophication, while summertime  $NO_x$  emissions reductions are most helpful in attaining the  $O_3$  standard. In some cases,  $PM_{10}$  nonattainment and visibility impacts are strongly related to seasonal conditions. Since regulatory programs may be designed to achieve emissions decreases at a constant, year-round rate or on a seasonal basis, the impacts of the policy decisions on the timing of the required decreases need to be understood and, to the extent possible, integrated.

# NO<sub>x</sub> Emissions by Season

As shown in the table at the end of this section, total NO<sub>x</sub> emissions vary somewhat by season, with summer emissions usually slightly highest (EPA, 1995). Several source categories emit evenly throughout the year. NO<sub>x</sub> emissions from electric utilities are highest in the summer. On-road NO<sub>x</sub> emissions vary little from season to season, but are slightly higher in the summer than winter. Non-road engine emissions (non-road diesel and gasoline) are higher in the summer than in the winter since weather limits the use of these engines for construction, lawn and garden, recreation, and light commercial purposes.

# Seasonal Considerations in Environmental Programs Acid Deposition

The impacts from acid deposition are both cumulative and short-term. The cumulative effects are due to long-term chronic acidification of watersheds. In addition, some important adverse effects are associated with springtime snowmelt. As described in section II.A of this document, nitric acid deposition plays a dominant role in the acid pulses associated with the fish kills observed during the springtime melt of the snowpack in sensitive watersheds. Thus, wintertime NO<sub>x</sub> emissions reductions are especially important to lessening the incidence and severity of acidic episodes in certain areas. In addition, the timing of aluminum concentration

peaks is also important. Toxic aluminum peaks related to nitrate fluctuations commonly occur in late summer or early fall when soil temperatures and root growth are usually high (Joslin et al., 1992). Continuous year-round  $NO_x$  emissions reductions appear to be the most beneficial for decreasing acid deposition damage to natural resources.

### Eutrophication

N is the limiting nutrient in most coastal estuaries and many lakes. Thus, as described in

section III.B of this document, addition of N results in accelerated plant growth in the waterbody causing adverse ecological effects and economic impacts that range from nuisance algal blooms to oxygen depletion and fish kills. Simulated water quality responses to year-round vs seasonal nutrient emissions reductions conducted as a 1992 reevaluation of the baywide Tributary Nutrient Reduction Strategy by the Chesapeake Bay Program indicated the need for year round controls on phosphorus and N loadings (Thomann et a. 1994).

#### Ground-level $O_3$

High ambient concentrations of  $O_3$  are associated with periods of elevated temperature and solar radiation. Thus, in most parts of the country, high  $O_3$  episodes occur only during summer months. Accordingly, the control of  $NO_x$  emissions on a summer season basis may be part of some areas' strategies to attain the  $O_3$  standard at least cost. It should also be noted that application of  $NO_x$  emissions controls that focus emissions reductions in the summer will, in many cases, also achieve significant emissions reductions on a year-round basis. For example, efforts to decrease emissions from large boilers will usually include installation of low  $NO_x$ burners--which will achieve year-round moderate amounts of emissions reductions--and may include, in addition, some type of summer season control, such as switching to a cleaner fuel or post-combustion technology. In some cases, year-round emissions reductions contained in States'  $O_3$  attainment plans are explicitly supplemented by seasonal requirements; e.g., the Northeast States adopted a strategy which calls for  $NO_x$  emissions reductions in three phases over the region with Phase 1 requiring year-round controls and the subsequent phases covering the May through September timeframe.

# $NO_2$

Since the NO<sub>2</sub> NAAQS requires ambient concentrations to be averaged over an annual period, seasonal emissions should not affect this program.

#### РМ

In some cases  $PM_{10}$  nonattainment is related to seasonal emissions. For example, in some mountain/valley locations, the burning of wood for heating purposes results in wintertime exceedances of the NAAQS. In other cases,  $PM_{10}$  nonattainment is related to a variety of sources on a non-seasonal basis.

# Visibility and Regional Haze

Visibility is lowest in the summer in the region south of the Great Lakes and east of the Mississippi; in some locations the light extinction is more than twice as great in the summer as during the other seasons (NRC, 1993). The most intense regional haze in the US occurs in the east, where summertime meteorological conditions associated with slow-moving high-pressure systems create stagnant conditions (NRC, 1993). Thus, in the summer, pollutants from many different sources can accumulate, causing severe and widespread visibility degradation.

In the Grand CanyonVisibility Transport Commission's June 10, 1996 report it is noted that seasons influence the relative visibility impacts of regional and local emissions. Both emission types contribute to visibility impairment much of the year, and either type can be the

dominant cause of impairment on any particular day regardless of the season.

# Example of Consideration of Seasonal Concerns in $O_3$ Policy

# $O_3$ Season Emissions Limit

To help achieve the  $O_3$  standard, many areas require application of RACT on major stationary sources of  $NO_x$ . Typically these controls involve modifications to combustion equipment, such as installation of low  $NO_x$  burners. Such controls operate continuously, year-round.

EPA guidance issued in 1993 (Shapiro, 1993) gives States the option to allow sources to control  $NO_x$  emissions by switching to cleaner fuels during the  $O_3$  season. The  $NO_x$  emissions reductions must be equal to or greater than emissions reductions that would have occurred from application of typical RACT controls. In general, a fuel-switching program would provide new flexibility to States and industry in meeting certain Act requirements, including the  $NO_x$  RACT requirements. Fuel switching is a viable option for units where natural gas is readily available since the price of natural gas in the  $O_3$  season may be competitive with other fuels.

# Fuel-Switching Environmental Considerations

The EPA considered the relative environmental benefits for fuel switching and presumptive NO<sub>x</sub> RACT. In terms of the primary purpose of NO<sub>x</sub> RACT, that is lowering O<sub>3</sub> effects in areas of high concentrations, it is clear that the NO<sub>x</sub> emissions reductions due to burning a cleaner fuel during the O<sub>3</sub> season would be much more effective than lesser emissions reductions at the presumptive NO<sub>x</sub> RACT levels, which would be evenly spread over an entire year. The use of natural gas instead of coal could also substantially decrease annual and summertime emissions of SO<sub>2</sub>, carbon dioxide (CO<sub>2</sub>), PM<sub>10</sub>, and associated toxic emissions such as mercury. Further, emissions reductions of these pollutants may be especially effective in the summer with respect to decreasing regional haze and sulfate-related PM, both of which tend to peak in the summer. Thus, the potential benefits that go beyond the title I O<sub>3</sub> and NO<sub>x</sub> RACT goals include helping attain/maintain the NAAQS for SO<sub>2</sub> and PM, decreasing mercury and other air toxic emissions, improving visibility, and cutting emissions of CO<sub>2</sub>, a global warming gas.

The EPA also considered evidence suggesting that, for certain ecosystems, decreases in N deposition that occur only during the summer would be less effective at decreasing acid deposition and nutrient impacts than emissions reductions that occur more uniformly throughout the year. It is not possible at this time to determine or fully quantify this relative ecological impact. Moreover, due to the inherent limits on the amount of fuel switching that can occur and the required NO<sub>x</sub> emissions reductions under the CAA, wintertime N deposition would be projected to decrease in most areas regardless of fuel switching. In contrast, the O<sub>3</sub> related benefits--and many of the additional potential benefits of fuel switching noted above--are well known and quantifiable. In conclusion, it was EPA's judgment that substantial decreases in O<sub>3</sub> concentrations would occur from fuel switching; this benefit and the accompanying improvements in visibility, PM, air toxics, and global warming that also occur from fuel switching clearly outweigh the decreased year-round benefits.

Source Category	1985	1986	(thousand 1987	1988	1989	1990	1991	1992	1993	1994
Fuel CombElectric Utility	6,916	6,909	7,128	7,530	7,607	7,516	7,488	7,475	7,773	7,795
Winter	1,801	1,757	1,748	1,928	1,980	1,804	1,858	1,864	1,914	1,913
Spring	1,603	1,568	1,627	1,679	1,300	1,729	1,722	1,768	1,779	1,772
Summer	1,862	1,935	2,055	2,137	2,034	2,104	2,064	2,019	2,188	2,207
Fall	1,650	1,648	1,698	1,786	1,833	1,879	1,843	1,825	1,893	1,904
Fuel CombIndustrial	3,209	3,065	3,063	3,187	3,209	3,256	3,175	3,216	3,197	3,206
Winter	825	789	788	820	825	837	816	827	822	825
Spring	794	758	757	789	794	806	786	796	791	793
Summer	785	749	748	779	784	796	777	786	782	784
Fall	805	769	769	800	805	817	796	806	802	804
Fuel CombOther	712	694	706	740	736	712	719	730	726	727
Winter	343	328	332	349	349	334	338	344	341	343
Spring	163	162	165	172	172	167	167	171	170	170
Summer	56	56	59	61	59	59	59	60	60	60
Fall	149	148	151	158	155	151	155	156	155	154
Chemical & Allied Product Mfg.	262	264	255	274	273	276	278	284	286	291
Winter	65	66	64	69	69	71	71	73	73	74
Spring	72	72	70	75	74	75	75	77	77	79
Summer	62	62	60	64	64	65	65	66	67	68
Fall	63	63	61	66	66	66	67	68	69	70
Metals Processing	87	80	75	82	83	81	78	80	81	84
Winter	22	20	19	20	21	20	19	20	20	21
Spring	22	21	19	21	21	21	20	20	21	22
Summer	21	20	18	20	20	20	19	20	20	21
Fall	21	20	19	20	21	20	19	20	20	21
Petroleum & Related Industries	124	109	101	100	97	100	97	96	95	95
Winter	31	27	25	25	24	25	24	24	23	23
Spring	31	27	25	25	24	25	24	24	24	24
Summer	31	27	25	25	24	25	24	24	24	24
Fall	31	27	25	25	24	25	24	24	24	24
Other Industrial Processes	327	328	320	315	311	306	297	305	315	328
Winter	79	79	77	76	75	74	72	74	76	79
Spring	83	83	81	80	79	77	75	77	80	83
Summer	83	83	81	80	79	77	75	77	80	83
Fall	82	83	81	79	78	77	75	77	79	83
Solvent Utilization	2	3	3	3	3	2	2	3	3	3
Winter	1 1	1 1	1 1	1 1	1 1	1	1 1	1	1 1	1
Spring	1		1	1		1	1	1		
Summer Fall		1			1 1	-		1	1	1
	1 2	1 2	1 2	1 2	2	1 2	1 2	1 3	1 3	1 3
Storage & Transport Winter	1	1	1	1	1	1	1	3 1	3 1	1
	1	1	1	1	1	1	1	1	1	1
Spring Summer	1	1	1	1	1	1	1	1	1	1
Fall	1	1	1	1	1	1	1	1	1	1
Waste Disposal & Recycling	87	87	85	85	84	82	83	83	84	85
Winter	21	22	21	21	21	20	20	21	21	21
Spring	22	22	21	21	21	20	20	21	21	21
Summer	22	22	21	21	21	21	21	21	21	21
Fall	22	22	21	21	21	21	21	21	21	21
On-Road Vehicles	8,089	7,773	7,651	7,661	7,682	7,488	7,373	7,440	7,510	7,530
Winter	2,063	1,962	1,938	1,949	1,956	1,881	1,863	1,873	1,907	1,925
Spring	2,000	1,974	1,939	1,942	1,938	1,907	1,857	1,880	1,896	1,897
Summer	2,040	1,920	1,885	1,879	1,903	1,864	1,834	1,855	1,856	1,866
Fall	1,983	1,918	1,889	1,892	1,885	1,836	1,820	1,833	1,851	1,841
Non-Road Sources	<b>2,734</b>	2,777	2,664	2,914	2,844	2,843	2,796	2,885	2,985	3,095
Winter	587	596	574	625	610	610	600	618	638	660
Spring	674	685	657	720	704	704	691	713	737	764
Summer	780	793	759	830	810	809	796	822	852	886
Fall	693	703	674	738	720	720	709	732	758	786
Miscellaneous	309	257	351	726	292	373	283	249	219	374
Winter	25	22	27	46	24	28	24	22	20	28
Spring	122	120	125	146	123	127	123	121	120	128
Summer	72	46	92	278	62	103	57	40	25	103
Fall	91	70	107	256	83	115	79	66	53	116
Total All Sources	22,860	22,348	22,403	23,618	23,222	23,038	22,672	22,847	23,276	23.615
Winter	5,861	5,668	5,615	5,928	5,955	<b>23,030</b> 5,747	5,705	5,757	5,856	5,910
Spring	5,624	5,485	5,483	5,665	5,704	5,761	5,572	5,680	5,717	5,760
Summer	5,775	5,718	5,807	6,180	5,864	5,830	5,797	5,796	5,976	6,128
Fall	5,599	5,476	5,499	5,845	5,700	5,700	5,597	5,613	5,725	5,817

# References

Grand Canyon Visibility Transport Commission, *Recommendations for Improving Western Vistas*, June 10, 1996 report of the Grand CanyonVisibility Transport Commission to the United States Environmental Protection Agency, June 10, 1996.

National Research Council, Committee on Haze in National Parks and Wilderness Areas, *Protecting Visibility in National Parks and Wilderness Areas*, 1993.

Shapiro, Acting Assistant Administrator, Office of Air and Radiation, U.S. Environmental Protection Agency, "Fuel Switching to Meet the Reasonably Available Control Technology (RACT) Requirements for Nitrogen Oxides (NO<sub>x</sub>)" EPA memorandum to Regional Air Directors, July 30, 1993.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *National Air Pollutant Emission Trends, 1900-1994*, October 1995, EPA-454/R-95-011.

C. Interface with Other Control Programs: Three Examples of Secondary Emissions, EPA's Clean Air Power Initiative, and New Standards for O<sub>3</sub> and Particulate Matter

# 1. Synthetic Organic Chemical Manufacturing Industry (SOCMI)

The CAA amendments of 1990 mandate that SIPs for certain  $O_3$  nonattainment areas be revised to require RACT to limit emissions of VOCs from sources for which EPA has published a control techniques guideline (CTG) document. Each CTG document contains a recommended "presumptive norm" for RACT for a particular source category, based on the EPA's current evaluation of capabilities and problems general to the source category. In some cases, controls for VOCs could increase NO<sub>x</sub> emissions (secondary emissions). In EPA's development of the SOCMI CTG for Halogenated and Nonhalogenated Vent Streams (EPA, August 1993), this issue was addressed. The CTG illustrates eight optional levels of control, with the NO<sub>x</sub> secondary emissions listed. In most options the amount of secondary NO<sub>x</sub> emissions is extremely small compared to the decrease in VOC emissions. In addition, the CTG addresses the issue of secondary emissions as follows:

Another important consideration in applying RACT is emission of pollutants such as carbon monoxide and nitrogen oxides from combustion-based control devices. The potential consequences of emission from control devices are twofold. First, depending on the VOCs-to-NO<sub>x</sub> ratio in the ambient air, NO<sub>x</sub> emissions from control devices may cause more O<sub>3</sub> to be formed than could be eliminated through VOC emissions reductions. Second, emissions from control devices may be enough to trigger New Source Review. (Table 6-1 shows expected national emissions of NO<sub>x</sub> and, in parentheses, the maximum annual emissions of NO<sub>x</sub> at a single facility.) Whether the VOC emission decreases are worth the increase in other pollutants from the VOCs control device is highly dependent on air quality and meteorological conditions in each specific geographical area. Therefore, States may select a less stringent level of control as RACT based on these considerations.

# 2. Pollution Control Projects

The EPA issued guidance (Seitz, 1994) which addresses issues involving the EPA's new source review (NSR) rules and guidance concerning the exclusion from major NSR of pollution control projects at existing sources. The guidance assures that any increase in NO<sub>x</sub> emissions due to decreases in VOC emissions would be minimized and that certain PSD requirements are met. That is, a qualifying add-on control device may be considered a pollution control project and may be considered for an exclusion from parts of EPA's NSR rules. The permitting agency should: (1) verify that the NO<sub>x</sub> increase has been minimized to the extent practicable; (2) confirm (through modeling or other appropriate means) that the actual significant increase in NO<sub>x</sub> emissions does not violate the applicable NAAQS PSD increment, or adversely impact any Class I area; and (3)

apply all otherwise applicable SIP and minor source permitting requirements, including opportunity for public notice and comment.

# *3. Landfill Methane*

As the waste in a landfill decomposes, it breaks down to form landfill gases, such as methane, smog-causing VOCs, and air toxics, pollutants known or suspected of causing cancer and other serious health effects. Landfills are the largest anthropogenic source of methane emissions in the United States. Methane is a greenhouse gas that contributes to global warming. In the March 12, 1996 Federal Register, EPA issued final rules for municipal solid waste landfills which will achieve significant decreases in emissions of VOCs, air toxics and methane.

This section provides a discussion of the secondary air emissions associated with municipal solid waste (MSW) landfill control devices such as flares, boilers, gas turbines, and stationary internal combustion (IC) engines. These control techniques, except flares, use the energy content of the landfill gas to generate electricity or steam. At the same time, the burning of the landfill gas produces  $NO_x$  emissions. Consequently, EPA is concerned about the impact of these secondary emissions in evaluating the overall benefits of applying landfill air emission controls. The overall impact on  $NO_x$  emissions, however, appears to be a decrease (EPA, 1991).

In evaluating the options for control of air emissions at MSW landfills, it is important to consider the overall impact of the controls. The emission controls involving energy recovery generally yield electricity or steam. Thus, landfill energy recovery devices such as gas turbines and IC engines are expected to decrease local or regional electric utility power generation. The electricity or steam produced by these controls would otherwise be produced by some other means. This decrease in utility requirements is likely to result in the reduction of  $NO_x$  emissions from coal-fired power plants.

In this analysis, electricity generated from landfill energy recovery techniques is assumed to displace an equal amount of electricity that would otherwise be generated from coal-fired utility boilers. Based on current utility fuel costs, this is a reasonable assumption. Therefore, the net secondary air impacts represent the difference between air emissions generated by the control equipment and air emissions that would be generated from producing an equivalent amount of electricity with a coal-fired boiler/steam turbine.

The EPA judged that an analysis of secondary emissions from control techniques at MSW landfills should consider the differential between emissions from an IC engine or a gas turbine and the emissions they might "displace" at a coal-fired utility plant under the rules for coal-fired utility boilers (40 CFR 60, Subparts D and Da). The emission factors for the energy recovery techniques were simply compared to the emission factors for the utility boiler to estimate relative impacts. The EPA analysis (EPA, 1991) found that overall NO<sub>x</sub> emissions would be decreased at the following rates in cases where a gas turbine or IC engine is the control device: 224 and 139 pounds per million standard cubic feet of landfill gas, respectively.

# Clean Air Power Initiative

#### Summary

The Clean Air Power Initiative (CAPI) is a multi-stakeholder project intended to improve air pollution control efforts involving the power generating industry. The project's goal is to develop an integrated regulatory strategy for three major pollutants emitted from electric power generators; namely, SO<sub>2</sub>, NO<sub>x</sub>, and, potentially, mercury. Major decreases in these pollutants are expected to be needed to reduce the detrimental health effects of ground-level O<sub>3</sub>, fine particles, and hazardous air pollutants and reduce the environmental effects of acidification, eutrophication, ecosystem, crop, and materials damage, and regional haze. CAPI has considered, where feasible, new approaches to pollution control that recognize the long-range transport of many air pollutants and the economic benefits of emissions trading. The project was initiated by EPA's Assistant Administrator for Air and Radiation in 1995. As individual companies develop and implement strategies to participate in more competitive power markets, they could benefit from greater certainty in being able to plan for and reduce costs of future environmental regulations. The EPA is interested in reinventing its regulatory approach to decrease the number, administrative complexity, and cost of its requirements while improving the likelihood of achieving environmental results. The air quality improvement scenarios considered under the CAPI could be implemented through existing CAA authority, however new congressional authority may be preferable. A strategy, agreed to and supported by multiple stakeholders, would provide an opportunity to protect public health and the environment at lower cost to the power generation industry and to taxpayers than traditional regulation.

#### Background

Emissions from fossil-fuel-fired electric power plants contribute significantly to a number of important air pollution and multi-media issues. These can be briefly categorized as: 1) adverse effects on human health from ground level  $O_3$ , PM, and persistent toxic air contaminants; 2) environmental impacts such as eutrophication of coastal surface waters, wide-spread regional haze that decreases visibility, acidification of surface waters from acid deposition, ecosystem and crop damage from ground level  $O_3$ , and ecosystem damage from mercury and other persistent toxic pollutants; and 3) climate change due to greenhouse gases. In 1994, power plants were responsible for 70 percent of all sulfur oxide (SO<sub>x</sub>) emissions, 33 percent of all NO<sub>x</sub> emissions, 23 percent of point source emissions of direct or "primary" PM, 23 percent of anthropogenic mercury emissions, and 36 percent of all anthropogenic CO<sub>2</sub> emissions (EPA, 1995). In addition, power plants contribute to a range of other environmental impacts due to their water consumption and disposal of solid wastes.

For purposes of the CAPI, EPA has chosen to focus on the pollutants that are related to the first two categories of health and environmental effects noted above because these pollutants are associated with pressing regional health and environmental concerns in North America. In addition, EPA has clear statutory authority to regulate these pollutants. From a control/emissions perspective, the pollutants of greatest importance can be grouped into three categories: sulfur oxides,  $NO_x$ , and mercury and other directly emitted toxic fine particles. Sulfur and  $NO_x$  emissions undergo complex atmospheric transformations that result in the formation of acidic fine particles and gases,  $O_3$  smog, and toxic pollutants. The resultant mix, along with directly emitted mercury and fine particles, can be transported by weather systems over long distances and affect

air and water quality in areas far from where they were emitted. Because of this long range transport and the location of multiple power generation emission sources in the United States, the resulting atmospheric and deposition problems affect broad multi-state regions.

The CAA as amended in 1990 contains multiple requirements that will affect the power generating industry well into the future. Regulations based on some of these requirements have already taken effect and are being implemented, such as the  $SO_2$  allowance trading program under Title IV and  $NO_x$  RACT (an emissions standard based on RACT) under Title I. However, other mandated measures and requirements are still under development. One of the most difficult and important issues in this regard is related to developing  $O_3$  attainment plans under Title I. In addition, the CAA mandates that EPA periodically review standards and conduct studies that could affect the power generation industry over the next 10 years.

In implementing the CAA requirements, it is important to recognize that the electric power industry is facing major changes in the way it is structured and the way it generates, transmits, and distributes electricity. Competition is building in the industry in response to changes in the law, technology, and markets. In a non-competitive environment, electric utilities faced with pollution control requirements were allowed by their regulators to pass on environmental costs to consumers in the form of higher electricity rates. In a competitive market, utilities that have higher rates because of pollution controls would be at a relative disadvantage, while those with lower or no pollution control costs could increase market share. From an environmental perspective, there is concern that, absent appropriate emissions controls and policy instruments allowing for reduced compliance costs, the overall move towards a competitive market could result in significant regional environmental degradation (Browner, 1996).

Implementing the multiple requirements in a piecemeal fashion is unlikely to result in economically optimal results. Many of the individual actions would ultimately result in different source-specific emissions requirements or specific control technology mandates for the same pollutants. On this basis alone, it is worth examining coordination of the activities. However, our understanding of the nature of the environmental effects of power generation emissions suggests the possibility of significantly more efficient and effective approaches.

As noted above, the current environmental issues associated with power generation are related to regional scale transport of the emissions and transformation products of three key pollutants released from hundreds of sources. This decreases the need to be concerned about single source specific effects that are typically addressed with command and control approaches. A focus on regional emissions reductions enables us to consider regional market based solutions that have proven to be considerably more cost-effective. Conceptually, the multiple regional problems associated with these emissions could be addressed by establishing an emissions budget or cap at a set amount within a certain geographic area, allocating those emissions to sources, and allowing sources within that region to trade their emissions with one another ("cap and trade" approach). While the current experience with this approach has been with the Congressionally mandated Title IV acid rain program, the States and EPA have already made considerable

progress in developing emissions budget approaches for implementing regional  $NO_x$  control programs. Such approaches could also readily be considered for potential fine particle programs or regional persistent toxic pollutants such as mercury.

Beyond the consideration of market-based approaches, EPA believes it is highly desirable to engage key stakeholders in the industry, states, and environmental groups in a discussion of a more effective way to establish cost-effective implementation approaches for addressing the key air emissions from the power generating industry. Agreements on desirable interim emissions reduction targets for fixed time periods could provide regulatory certainty for the industry, assurances that emissions reductions would be achieved and sustained, more cost-effective emissions reductions for companies and their customers, and reduce the resources all parties must expend on issue-by-issue rule development, risk and cost assessments, control strategy plan development, permitting, monitoring, and litigation.

#### Goals of the Initiative

CAPI is intended to break the current combative and costly pattern of regulation with a new collaborative approach that assures the public of the health and environmental protections promised by the CAA while providing the power generation industry with more certainty of future regulatory requirements, greater flexibility, and cost savings. EPA believes consideration of a cap and trade approach for  $SO_2$ ,  $NO_x$ , and potentially, mercury (with appropriate local safeguards, such as Title IV provides) is appropriate because these pollutants are transported far from their source and much of the current health and environmental damage caused by these pollutants comes from their total loadings in the air and on the ground.

# Progress to Date

EPA began CAPI in 1995 by holding a series of small meetings with interested stakeholders and by developing a model that could analyze the cost and emission implications of different emissions reduction scenarios for  $SO_2$ ,  $NO_x$ , and mercury. Detailed information about the model and the various scenarios analyzed will not be presented in this paper, but are well documented elsewhere<sup>17</sup>. In 1996, EPA held public meetings in April, May, and July to continue and expand the dialogue on CAPI and to hear reactions to various emissions reduction scenarios that EPA believes are consistent with the requirements of the CAA. As described in EPA's CAPI report (EPA, October 1996) EPA also believes that cap and trade programs are effective ways to ensure that environmental goals are maintained in the future without continually returning to the industry for more emissions reduction actions. EPA will continue to improve the modeling tool developed for CAPI, test other scenarios and sensitivity cases, explore economic and environmental impacts, and provide input to the Clean Air Act Advisory Committee deliberations. EPA would also like to continue to work with the power generation industry to find cost-effective solutions to our environmental problems

<sup>&</sup>lt;sup>17</sup> See 1) <u>EPA's Forecast of Electric Power Generation and Air Emissions</u>, and 2) <u>Analysis of Options for Air</u> <u>Emissions Control Under the Clean Air Power Initiative</u>, Office of Air and Radiation, U.S. EPA April 1996; and 3) <u>Revised Forecast of Electric Power Generation</u>; 4) <u>Analyzing Electric Power Generation Under the CAAA</u>, July 1996; and 5) <u>Supporting Analysis for EPA's Clean Air Power Initiative</u>, Office of Air and Radiation, U.S. EPA, October 1996.

# New Standards for $O_3$ and PM Common Factors for $O_3$ and PM

As described in the sections on "Ozone" and "Particulate Matter," EPA published revisions to the  $O_3$  and PM NAAQS on July 18, 1997 (62 FR 38856). As part of the revisions process, EPA initiated action to address strategies for the implementation of the new NAAQS. These ongoing reviews and related implementation strategy activities to date have brought out important common factors between  $O_3$  and PM. Several similar health effects have been associated with exposure to  $O_3$  and PM, including for example aggravation of respiratory disease (e.g., asthma), increased respiratory symptoms, and increased hospital admissions and emergency room visits for respiratory causes.

Other similarities in pollutant sources, formation, and control exist between O<sub>3</sub> and PM, in particular the fine fraction of particles. These similarities include: (1) atmospheric residence times of several days, leading to regional-scale transport of the pollutants; (2) similar gaseous precursors, including NO<sub>x</sub> and VOCs, which contribute to the formation of both O<sub>3</sub> and PM in the atmosphere; (3) similar combustion-related source categories, such as coal and oil-fired power generation and industrial boilers and mobile sources, which emit particles directly as well as gaseous precursors of particles (e.g., SO<sub>x</sub>, NO<sub>x</sub>, VOCs) and O<sub>3</sub> (e.g., NO<sub>x</sub>, VOCs); and (4) similar atmospheric chemistry driven by the same chemical reactions and intermediate chemical species which favor both high  $O_3$  and fine particle concentrations. High fine particle concentrations are also associated with significant impairment of visibility on a regional scale. These similarities provide opportunities for optimizing technical analysis tools (i.e., monitoring networks, emissions inventories, air quality models) and integrated emission reduction strategies to yield important cobenefits across various air quality management programs. This integration could result in a net reduction of the regulatory burden on some source category sectors that would otherwise be impacted separately by O<sub>3</sub>, PM, and visibility protection control strategies. In recognition of the potential benefits of integrating the Agency's approaches to providing for appropriate protection of public health and welfare from exposure to O<sub>3</sub> and PM, the Agency plans to develop associated implementation strategies under coordinated schedules.

# Integrated Implementation of the New O<sub>3</sub> and PM Standards

The EPA initiated a process designed to provide for significant stakeholder involvement in the development of integrated implementation strategies for the new/revised  $O_3$  and PM NAAQS and a new regional haze program. As described below, this process involves a new subcommittee of the Agency's Clean Air Act Advisory Committee (CAAAC), established in accordance with the Federal Advisory Committee Act (FACA) (5 U.S.C. App.2).

The FACA was enacted in 1972 to open the advisory committee process to public scrutiny and to protect against undue influence by special interest groups over government decision making. Federal Advisory Committees may be established by statute, the President, or by the head of a Federal Agency. An advisory committee or subcommittee is established under FACA to obtain advice or recommendations from advisory groups established by or closely tied to the Federal Government. The CAAAC was established to provide independent advice and counsel to the EPA on policy and technical issues associated with the implementation of the Act. The CAAAC advises EPA on the development, implementation, and enforcement of several of the new and expanded regulatory and market-based programs required by the Act.

The CAAAC advises on issues that cut across several program areas. The programs falling under the purview of the CAAAC include those for meeting NAAQS, reducing emissions from vehicles and vehicle fuels, decreasing air toxic emissions, issuing operating permits and collecting fees, and carrying out new and expanded compliance authorities. The CAAAC holds meetings, analyzes issues, conducts reviews, performs studies, produces reports, makes recommendations, and undertakes other activities necessary to meet its responsibilities. Comments, evaluations, and recommendations of the CAAAC and responses from the EPA are made available for public review, in accordance with Section 10 of FACA.

A new subcommittee of the CAAAC, the Subcommittee for Ozone, Particulate Matter, and Regional Haze Implementation Programs (the Subcommittee), was established in August 1995 to address integrated strategies for the implementation of potential new  $O_3$  and PM NAAQS, as well as a regional haze program. The Subcommittee is composed of representatives selected from among state, local, and tribal organizations; environmental groups; industry; consultants; science/academia; and federal agencies. Recommendations made by the Subcommittee will be submitted to EPA through CAAAC. To facilitate communication between the Subcommittee and CAAAC, some members of CAAAC are on the Subcommittee.

The Subcommittee is charged with providing advice and recommendations to EPA on developing new, integrated approaches for implementing potential revised NAAQS for  $O_3$  and PM, as well as for implementing a new regional haze reduction program. The Subcommittee is expected to examine key aspects of the implementation programs for  $O_3$  and PM, to provide for more flexible and cost-effective implementation strategies, as well as to provide new approaches that could integrate broad regional and national control strategies with more localized efforts. In addition, the Subcommittee will consider new and innovative approaches to implementation including market-based incentives. The focus of the Subcommittee will be on assisting EPA in developing implementation control strategies, preparing supporting analyses, and identifying and resolving impediments to the adoption of the resulting programs.

Issues involved in the revision of the  $O_3$  and PM NAAQS, such as the averaging time, level, and form of any revised standards, were addressed in accordance with the NAAQS review process described in the above sections, including review by CASAC, and are not within the Subcommittee's charge. CASAC is charged with providing advice and recommendations to the Administrator on all matters pertaining to the review of and possible revisions to the NAAQS. Similarly, selection of the appropriate indicator or units of measurement for quantifiable changes in visibility are being addressed through an independent, scientific peer-review process and, thus, will not be a subject for recommendations by the Subcommittee.

References

Browner, C. M., Administrator, U.S. Environmental Protection Agency, letter to Council on Environmental Quality, May 13, 1996.

Seitz, John, Director, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, "Pollution Control Projects and New Source Review (NSR) Applicability," July 1, 1994 memorandum to Regional Air Directors.

U.S. Environmental Protection Agency, Office of Air and Radiation, "EPA's Clean Air Power Initiative" October 1996.

U.S. Environmental Protection Agency, Office of Air and Radiation, "EPA's Forecast of Electric Power Generation and Air Emissions," and "Analyzing Electric Power Generation Under the CAAA," April 1996.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *Control of Volatile Organic Compound Emissions from Reactor Processes and Distillation Operations Processes in the Synthetic Organic Chemical Manufacturing Industry*, August 1993, EPA-450/4-91-031.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *Air Emissions from Municipal Solid Waste Landfills, Background Information for Proposed Standards and Guidelines*, EPA-450/3-90-011a, March 1991.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *National Air Pollutant Emission Trends*, 1900-1994, October 1995, EPA-454/R-95-011.

# Appendices

# Introduction

Appendices A and B provide information on specific programs currently underway to achieve decreases in emissions of  $NO_x$ . In some cases, these are programs specifically mandated by the CAA. In other cases, they are programs that are needed to meet more general CAA requirements. In addition, several related efforts by the States are described. These are control programs that affect both new and old sources of emissions and are generally based on currently available control technologies.

Since passage of the 1970 CAA amendments, air pollution control and prevention technologies have continuously improved. Technologies such as selective catalytic reduction and gas reburn systems are in place and successfully performing today that were only on the drawing board ten years ago. As the demand for more innovative and cost-effective or cost-saving technologies increases--due to the above new initiatives, for example--new technologies such as ultra low-NO<sub>x</sub> gas-fired burners and vacuum insulated catalytic converters will move from the research and development or pilot program phase to commercial availability. Thus, it is likely that many new technologies will be available in the next ten to fifteen years to employ in air pollution control and prevention strategies.

Appendix C provides more detailed information with respect to sources of  $NO_x$  emissions. In addition, Appendix C describes mechanisms that eventually remove  $NO_x$  from the atmosphere and provides some information on emissions of ammonia. Finally, Appendix D lists several acronyms and abbreviations.

# A. Mobile Source Programs

The control of  $NO_x$  emissions from mobile sources is under much more investigation since the CAA amendments of 1990. Light-duty vehicles, heavy-duty engines, nonroad engines, and fuels are all required to produce lower emissions. However, the United States has experienced tremendous growth in the activity of mobile sources overwhelming much of the emissions reductions from the introduction of cleaner technologies; vehicle miles traveled has grown exponentially without signs of leveling since the passage of the initial Clean Air Act.

#### Light-Duty Vehicles

# Tailpipe Standards

Programs that decrease or can decrease  $NO_x$  emissions from light-duty vehicles are numerous and involve all facets of the operation of these vehicles. These range from performance requirements like tailpipe standards to fuel modifications to in-use repairs.

The tailpipe standards for light-duty vehicles are evolving as a result of the 1990 CAA amendments and California rulemakings. The CAA established more stringent Tier 1 tailpipe emission standards, lowering the NO<sub>x</sub> emissions standard from 1.0 gram per mile to 0.4 grams per mile. In addition, states were allowed to adopt California Low Emission Vehicle (LEV) standards which further increased the stringency of the NO<sub>x</sub> standard. This led to a discussion about a new emission control program called the National Low Emission Vehicle (NLEV.) program, a voluntary but enforceable program which included a 0.2 gram per mile NO<sub>x</sub> standard for light-duty vehicles. The CAA also requires that EPA conduct a study to determine whether more stringent tailpipe standards (called "Tier 2" standards) are necessary, technically feasible, and cost effective. This study is currently underway.

EPA has recently updated the modeling of emission benefits that would result from the NLEV program. Based on a 1999 model year program start in northeast states and a 2001 model year start nationwide, as well as on realistic assumptions regarding individual state adoptions of the California LEV program, EPA estimates that  $NO_x$  will be reduced by 538 tons per day nationwide in 2005 and 1699 tons per day when the program is fully implemented in 2015.

Based on a detailed assessment by California in 1994 and updated in April, 1996, California has estimated the incremental cost of a LEV in California to be \$120 per car. EPA believes that the incremental cost for NLEV will be lower than the California estimate due to technology advances, harmonization of California and federal programs, economies of scale resulting from a nationwide program, and historical evidence that suggests that California's own cost estimates have generally been overstated and that the prices of newly-introduced technologies often decrease in successive model years.

#### Tailpipe Standards Summary

Reductions: Tier 1 sets a standard intended to achieve a 60% NO<sub>x</sub> emissions reduction in grams per mile (phasing in 1994-1996); NLEV could achieve an additional 50% NO<sub>x</sub> emissions reduction in grams per mile.
 Reference: NLEV Proposed Rulemaking Notice of 10-10-95; Final Rulemaking 6-7-97 (62 FR 31192).

#### **Onboard Diagnostics**

Another requirement of the CAA was the incorporation of On-board Diagnostic systems to determine the functionality of the emission control devices. This strategy might lower the cost of in-use compliance by notifying operators and repair personnel of problems with the emission control devices. The benefits and costs of the program depend in large measure on the emission standards imposed on the design of new vehicles. It also depends on the willingness or requirements of owner/operators to repair detected malfunctions. It is expected that  $NO_x$  emissions reductions would be derived from the installation of sensors to detect failures of the engine to meet its emission standards. A notice of proposed rulemaking was issued on May 28, 1997, proposing changes to the existing regulations.

# Supplemental Federal Test Procedure

The CAA also required that EPA reexamine the test procedure for light-duty vehicles.

Following an extensive study of in-use driving behavior and testing of in-use emissions, EPA completed a rule in October 1996 that supplements the existing test procedure to include higher speeds, higher acceleration driving behavior, and the operation of the vehicle air conditioner. EPA found that vehicles produce more  $NO_x$  emissions when operated under the higher loads, particularly with the air conditioner operating. Due to the change in the test procedure, vehicle emissions will need to be decreased to meet the current emissions limits for light-duty vehicles. That is, the more stringent supplemental test procedure is expected to result in reductions in VOCs, CO, and  $NO_x$  emissions;  $NO_x$  emissions are expected to be lowered by roughly 10% which, depending upon the future tailpipe standards, can produce  $NO_x$  emission reductions of about 125,000 to 400,000 tons. The cost of such a program is uncertain but EPA estimates that the cost effectiveness of the institution of a new standard is about \$1,000 per ton of VOC and  $NO_x$  emissions decreased. For further information, EPA published a final rule in the Federal Register on October 22, 1996 (61 FR 54851).

# Supplemental Federal Test Procedure Summary

- Reductions: 125,000-400,000 tons per year or roughly 10%.
- Reference: Final Rulemaking Notice of October 22, 1996.

# Clean Fuel Fleets

Provisions of the CAA amendments of 1990 require the establishment of a clean-fuel fleet program in certain  $O_3$  and CO nonattainment areas. This requires that some of the new vehicles purchased by certain fleet owners meet clean-fuel fleet vehicle exhaust standards. These requirements apply to light-duty vehicles and trucks and heavy-duty engines. The most recent rulemaking for this program was published in the <u>Federal Register</u> on September 30, 1994.

#### Transportation Alternatives

While CAA section 108(f) lists transportation control measures that may lower vehicle miles traveled (VMT), EPA does not require states or local areas to adopt specific transportation control measures. The CAA simply requires severe and extreme  $O_3$  areas to adopt "transportation control strategies and transportation control measures to offset any growth in emissions due to growth in VMT and numbers of vehicle trips," and to implement transportation control measures as necessary for attainment. EPA has interpreted the offset requirement as only applying to emissions of VOCs but it might have some effect on NO<sub>x</sub> emissions. EPA has also issued guidance for states to use in calculating activity growth in order to determine its scope and trend, "Section 187, VMT Forecasting and Tracking Guidance", January, 1992. The guidance is meant to ensure that growth will be properly determined and monitored.

#### In-Use Initiatives for Light-Duty Vehicles

Due to poor maintenance or deliberate tampering, motor vehicles in use have consistently emitted pollutants in excess of the established standards. Motor vehicle Inspection and Maintenance (I/M) programs have been singled out as the primary means to rectify these problems by identifying vehicles in need of repair. Many areas are required to implement I/M programs with various stringency to control emissions of CO, VOCs, and NO<sub>x</sub>. The benefits of enhanced I/M programs on NO<sub>x</sub> emissions is estimated to be about 9% with overall tonnage

reductions dependent upon the stringency of the tailpipe standards, possibly approaching 500,000 tons per year with wide coverage. The cost effectiveness of an I/M program targeted solely for  $NO_x$  emission reductions has not been determined. The final rule for I/M programs was published on November 5, 1992.

# Inspection/Maintenance Summary

- Reductions: 9%NO<sub>x</sub> reduction; up to 500,000 tons per year.
- Reference: Final Rulemaking Notice of 11-5-92 (57 FR 52950).

# Highway Heavy-Duty Vehicles

New heavy-duty engines intended for use in highway vehicles have been required to meet more stringent  $NO_x$  emission limits for engines built in 1985, 1990, and 1991 with subsequent reductions in those limits for 1998. While it is estimated that emissions per engine-mile will be significantly reduced once fleet turnover fully implements the new technologies, growth in activity will largely overwhelm these reductions. EPA has proposed new standards to begin with new engine manufactured in 2004 that would reduce  $NO_x$  emissions by over 1,000,000 tons by the year 2020. The cost effectiveness of the program is estimated to be \$200 to \$500 per ton of  $NO_x$ emissions decreased.

#### Heavy-Duty Vehicles Summary

- Reductions: 50% Reduction from 1998 NO<sub>x</sub> Levels by 2020.

- Reference: Notice of Proposed Rulemaking on 6-27-96 (61 FR 33469). Final Rulemaking expected in the fall of 1997.

# Nonroad Engines

There are several nonroad engine control programs some of which target VOC emissions, and some which target  $NO_x$  emissions. Those controls that target  $NO_x$  emissions are for large marine, aircraft, locomotive and general purpose engines like those used in agriculture, construction, and general industrial equipment.

# General Purpose Nonroad Engines

These engines produce the greatest portion of the nonroad NO<sub>x</sub> emissions. The EPA finalized an initial rule for compression-ignition (diesel) engines with rated power of over 37 kilowatts on June 17, 1994. The rule sets emission standards for new engines built starting in the years ranging from 1996 through 2000 depending upon the size of the engine. The cost effectiveness for this program was estimated at less than \$200 per ton of NO<sub>x</sub> decreased. The overall NO<sub>x</sub> emissions decreased once the rule is fully phased in, including engine turnover, is estimated at approximately 300,000 tons per year.

EPA intends to propose second and third tiers of standards for nonroad diesels in the fall of 1997. It is estimated that emission reductions for nonroad engines when the new standards are fully implemented will result in an additional reduction of 1,600,000 tons of NO<sub>x</sub> per year. EPA expects this program to be very cost effective, with cost per ton of NO<sub>x</sub> removed of under \$1000.

Reductions: In Phase I, a 20% reduction will produce about 300,000 tons per year reduction; Phases II and III could provide about an additional 40% reduction or about 800,000 tons per year.
Reference: Final Rulemaking Notice of 6-17-94; Advanced Notice of Proposed Rulemaking, August 31, 1995. EPA expects to formally propose emission limits for in the fall of 1997.

# Locomotive

The EPA proposed regulations for controlling locomotive emissions in January, 1997. This category represents the second largest producer of nonroad NO<sub>x</sub> emissions. The EPA is under a court-ordered deadline to complete a final rule by December 17, 1997. The rule as proposed on February 11, 1997 (62 FR 6365) includes stringent emission standards for all locomotives produced after the effective date of the standards, as well as requiring lesser emission reductions of most of the existing fleet of locomotives. The EPA estimates that standards under consideration will produce almost 400,000 tons of NO<sub>x</sub> emissions reduction per year by 2010, with ultimate reductions of almost 600,000 tons per year by 2040. The cost effectiveness of the control strategy is estimated in the proposal to be around \$175 per ton of NO<sub>x</sub> decreased.

#### Marine

The EPA is working with the International Marine Organization, a subgroup of the United Nations, to develop an agreement to control emissions from ships on international voyages. Such an agreement would provide important measures to control emissions from ships for which national standards could not apply. However at this time, the level and cost effectiveness of control measures have not been determined nor has there been any agreement.

#### Aircraft

The EPA is working with several government agencies and governments in the International Civil Aviation Organization to incorporate  $NO_x$  control measures for aircraft engines in an international agreement. However at this time, the level and cost effectiveness of control measures have not been determined nor has there been any agreement.

#### Fuels Programs

#### Gasoline

Reformulated gasoline (RFG) was instituted as a cleaner gasoline and was intended primarily as a VOCs emissions reduction strategy with a no  $NO_x$  increase requirement. The program is implemented in two phases with increasing stringency of the standard. Phase I provides for a decrease in VOCs and toxic emissions with a small (about 1.5%) decrease in  $NO_x$ emissions to provide a compliance margin for the no  $NO_x$  increase provision at minimal additional cost. In phase II of the program, EPA increased the stringency of VOCs control and exercised its discretion by imposing a  $NO_x$  control limit.

Phase II will include a requirement for a 6.8%  $NO_x$  control for the summer of the year 2000 in addition to more stringent VOC emissions reductions. The  $NO_x$  requirement will probably be achieved by lowering the amount of sulfur in gasoline. The standard was set on the basis of a marginal cost effectiveness, but the average cost effectiveness for the  $NO_x$  control standard is now projected to be less than \$2,000 per ton decreased. The overall  $NO_x$  emissions

reduction depends upon the amount of fuel that RFG displaces in the field, but for the nine cities required to implement RFG 22,000 summer tons of  $NO_x$  per year will be decreased. The program is mandatory for nine metropolitan areas and optional for other areas. Under certain conditions, states have the ability to mandate their own clean gasoline programs and are investigating those options. The final RFG rule was published in the Federal Register on February 16, 1994 (59 FR 7716).

#### RFG, Phase II Summary

- Reductions: Roughly 7% in areas used reducing nationwide  $NO_x$  emissions by at least 22,000 tons per summer season.

- Reference: Final Rulemaking Notice of February 16, 1994 (59 FR 7716).

#### Other Programs

#### Ozone Transport Assessment Group

The OTAG was a consultative process among the eastern States and EPA. The OTAG process assessed national and regional control strategies, using improved modeling techniques. Significant new modeling analyses were conducted by EPA and other agencies as part of the OTAG process. The goal of the OTAG process is for EPA and the affected States to reach consensus on the additional regional and national emission reductions that are needed to help achieve attainment of the  $O_3$  standard. On July 8, 1997 OTAG forwarded its final recommendations to EPA. Based on the results of the OTAG process, States are expected to submit in 1997 attainment plans which show attainment through local, regional and national controls.

#### OTAG Summary

Reductions: Depends on subsequent SIP revisions.

#### B. Stationary Source Programs

#### Emission Standards for Coal-Fired Power Plants

Title IV (Acid Deposition Control) of the CAA specifies a two-stage program for decreasing  $NO_x$  emissions from existing coal-fired electric utility power plants. Analogous to the national allowance program for decreasing  $SO_2$  emissions, this program is to be implemented in two phases. Phase I affected units (277 boilers) are required to meet the applicable annual emission rates beginning with calendar year 1996; Phase II affected units (775 boilers) are required to meet the applicable annual emission rates beginning with calendar year 1996; Phase II affected units (775 boilers) are required to meet the applicable annual emission rates beginning with calendar year 2000. Implementation of the first stage of the program, promulgated April 13, 1995 (60 FR 18751), will decrease annual NO<sub>x</sub> emissions in the United States by over 400,000 tons per year between 1996 and 1999 (Phase I) and by approximately 1.17 million tons per year beginning in 2000 (Phase II). These decreases are achieved by applying low NO<sub>x</sub> burner (LNB) technology to dry bottom wall-fired boilers and tangentially fired boilers (Group 1).

The second stage of the program, promulgated December 19, 1996 (61 FR 67112), provides for additional annual NO<sub>x</sub> emissions reductions in the United States of approximately 0.89 million tons per year beginning in the year 2000 (Phase II). Taken together, the two stages provide for an overall decrease in annual NO<sub>x</sub> emissions reductions in the United States of approximately 2.06 million tons per year beginning in the year 2000. In the second stage of the title IV Program EPA has: (1) determined that more effective low NO<sub>x</sub> burner (LNB) technology is available to establish more stringent standards for Phase II, Group 1 boilers than those established for Phase I; and (2) established limitations for other boilers known as Group 2 (wet bottom boilers, cyclones, cell burner boilers, and vertically fired boilers), based on NO<sub>x</sub> control technologies that are comparable in cost to LNBs.

The total annual cost of this regulation to the electric utility industry is estimated at \$267 million, resulting in an overall cost-effectiveness of \$227 per ton of NO<sub>x</sub> removed. The final rule sets lower Group 1 emission limits and establishes emission limits for several other types of coal-fired boilers (Group 2) in Phase II. The annual cost of these additional reductions will be approximately \$200 million, at an average cost-effectiveness of \$229 per ton of NO<sub>x</sub> removed. By the year 2000, the Phase II NO<sub>x</sub> rule will achieve an additional decrease of 890,000 tons of NO<sub>x</sub> annually.

Coal-Fired Power Plants (Group I)

- Reductions: 400,000 tons per year, Phase I
- Reference: Final rule published in the Federal Register of April 13, 1995.

Coal-Fired Power Plants (Phase II, Groups I & II)

- Reduction: 2,060,000 tons NO<sub>x</sub> emissions reduction, Phases I & II.
- Reference: Phase II proposed rule published in the Federal Register of December 19, 1996.

#### Clean Air Power Initiative (CAPI)

CAPI is a multi-stakeholder project intended to improve air pollution control efforts involving the power generating industry. The project's goal is to develop an integrated regulatory strategy for three major pollutants emitted from electric power generators; namely,  $SO_2$ ,  $NO_x$ , and mercury. Major decreases in these pollutants are needed to decrease the detrimental health effects of ground-level  $O_3$ , fine particles, and hazardous air pollutants and decrease the environmental effects of acidification, eutrophication, ecosystem, crop, and materials damage, and regional haze.

#### Industrial Combustion Coordinated Rulemaking

The CAA requires regulation of air emissions from several categories of industrial combustion sources, including boilers, process heaters, waste incinerators, combustion turbines, and internal combustion engines. These combustion devices are used pervasively for energy generation and waste disposal in a wide variety of industries and commercial and institutional establishments. They combust fuels including oil, coal, natural gas, wood, and non-hazardous wastes. Both hazardous air pollutants and criteria pollutants are emitted. The industrial combustion regulations will affect thousands of sources nationwide, and will have significant environmental and health impacts and cost considerations.

The EPA plans to implement an Industrial Combustion Coordinated Rulemaking to develop recommendations for Federal air emission regulations that address the various combustion source categories and pollutants. Regulations will be developed under sections 112 and 129 of the CAA, as well as section 111. The overall goal of the Industrial Combustion Coordinated Rulemaking is to develop recommendations for a unified set of Federal air regulations that will maximize environmental and public health benefits in a flexible framework at a reasonable cost of compliance, within the constraints of the CAA.

#### International NO<sub>x</sub> Protocol

The United States signed the Nitrogen Oxides Protocol in Sophia, Bulgaria in 1988. The Protocol caps national  $NO_x$  emissions. For the United States, this means a cap at 1984 levels or about 23.2 million tons per year. The cap is to be achieved by the year 1994. The United States emissions in 1994 were about 23.6 million tons. Due to emission reductions mandated by the CAA,  $NO_x$  emissions are projected (EPA Trends Report, October, 1995) to fall to 20.5 million tons by the year 2000 and remain below 22 million tons through the year 2010.

#### NO<sub>x</sub> Protocol

- Reductions: 0.4 million ton reduction needed from 1994 level.
- Reference: Nitrogen Oxides Protocol in Sophia, Bulgaria in 1988.

#### Municipal Waste Combusters

Standards of performance for new municipal waste combustor (MWC) units and emission guidelines for existing MWC's implement sections 111 and 129 of the CAA. The standards and

guidelines apply to MWC units at plants with aggregate capacities to combust greater than 35 megagrams per day (Mg/day) (approximately 40 tons per day) of municipal solid waste (MSW) and require sources to achieve emission levels reflecting the maximum degree of reduction in emissions of air pollutants that the Administrator determined is achievable, taking into consideration the cost of achieving such emissions reduction, and any non-air-quality health and environmental impacts and energy requirements. The promulgated standards and guidelines establish emission levels for MWC organics (dioxins/furans), MWC metals (cadmium (Cd), lead (Pb), mercury (Hg), PM, and opacity), MWC acid gases [hydrogen chloride (HCl) and SO<sub>2</sub>], NO<sub>2</sub>, and MWC fugitive ash emissions.

As explained at proposal (59 FR 48198, September 20, 1994), the combination of SD/FF, GCP, and SNCR was the basis of the new source MACT floor for  $NO_x$ . These technologies remain the basis for the final  $NO_x$  MACT floor. The final standard (December 19, 1995 Federal Register) for MWC's at large plants is 180 ppmv (24-hour averaging period) for the first year of operation, and 150 ppmv (24-hour averaging period) thereafter. The final standards do not require  $NO_x$  control for MWC's at small plants.

The EPA intends to amend the MWC standards in August 1997. The amended standards and guidelines would apply to only MWCs units larger than 250 tons per day capacity. (The MWC regulations would no longer apply to MWC plants larger than 35 Mg/day. Note the amendments would change both (1) size (250 tpd vs 35 Mg/day) and (2) unit capacity vs aggregate plant capacity.)

#### New Source Performance Standards (NSPS) for Boilers

Pursuant to section 407(c) of the CAA, EPA has reviewed the emission standards for  $NO_x$  contained in the standards of performance for new electric utility steam generating units and industrial-commercial-institutional steam generating units.(this requirement covers three existing NSPS in 40 CFR part 60: 1) Subpart Da for Utilities, 2) Subpart Db for Industrial boilers and Subpart Dc for small boilers). The proposed changes to the existing standards for  $NO_x$  emissions reduce the numerical  $NO_x$  emission limits for both utility and industrial steam generating units to reflect the performance of best demonstrated technology. The proposal also changes the format of the revised  $NO_x$  emission limit for electric utility steam generating units to an output-based format to promote energy efficiency and pollution prevention.

The primary environmental impact resulting from the revised  $NO_x$  standards is reductions in the quantity of  $NO_x$  emitted from new steam generating units subject to the proposed revisions to the NSPS. Estimated baseline  $NO_x$  emissions from these new steam generating units are 39,500 Mg/year (43,600 tons/year) from utility steam generating units and 58,400 Mg/year (64,400 tons/year) from industrial steam generating units in the 5th year. The revised standards are projected to reduce baseline  $NO_x$  emissions by 23,000 Mg/year (25,800 tons/year) from utility steam generating units and 18,000 Mg/year (20,000 tons/year) from industrial steam generating units in the 5th year after proposal. This represents an approximate 42 percent reduction in the growth of  $NO_x$  emissions from new utility and industrial steam generating units subject to these

#### revised standards.

#### **Boilers NSPS**

- 25,800 ton NO<sub>x</sub> emissions reduction annually from utility steam generating units
- 20,000 ton NO<sub>x</sub> emissions reduction annually from industrial steam generating units
- Reference: Proposed rule in the July 9, 1997 Federal Register

#### New Source Review

In  $O_3$  nonattainment areas (and in an Ozone Transport Region) major new or modified sources are required to control emissions by the Lowest Achievable Emission Rate and to offset the new emissions. In attainment areas the sources are required to control emissions by Best Available Control Technology and are not required to offset the new emissions. These programs help control emissions due to economic growth.

#### *NO*<sub>2</sub> *NAAQS Rulemaking*

Currently, all areas of the United States are in attainment of the annual NO<sub>2</sub> NAAQS of 0.053 ppm. Through implementation of NO<sub>x</sub> emissions reductions related to acid deposition and attainment of the O<sub>3</sub> and PM NAAQS, it is likely that the NO<sub>2</sub> standard will continue to be attained throughout the nation in the foreseeable future.

#### O<sub>3</sub> Attainment Plans

The 1990 CAA amendments provide the framework for action by states and EPA for national, regional, and local controls to achieve the NAAQS. Under these provisions, states are expected to submit SIPs demonstrating how each nonattainment area will reach attainment of the  $O_3$  NAAQS. Based on the degree that  $O_3$  concentrations in an area exceed the standard, the Act spells out specific requirements that states must incorporate into their attainment plans and sets specific dates by which nonattainment areas must reach attainment.

To help attain the 1-hour  $O_3$  air quality standard in the near-term, certain existing sources must install RACT and new sources must install controls representing the lowest achievable emission rate. To decrease acid deposition, the CAA requires coal-fired utility boilers to meet emission limits in two phases. Further, in a longer-term provision, the CAA requires States to adopt additional control measures as needed to attain the  $O_3$  standard. This requirement supplements the RACT and NSR requirements. Thus, a State would need to require NO<sub>X</sub> controls which achieve emission reductions greater than the NO<sub>x</sub> RACT/phase I acid deposition limits where additional reductions in emissions of NO<sub>x</sub> are necessary to attain the  $O_3$  standard by the attainment deadline.

The stationary (and mobile) source control measures needed for attainment will vary from region to region. For example, stationary sources in the South Coast Air Quality Management District of California are required to comply with Best Available Retrofit Control Technology (BARCT), which are stricter that Federal requirements. The application of BARCT to industrial sources generally results in  $NO_x$  emission rate reductions of 70-90 percent from uncontrolled levels.

#### Attainment Plans

- Reductions: Depending on (future) modeling findings and SIP revisions.

#### OTAG

Over a 2 year period EPA worked with OTAG, which was chartered by the Environmental Council of States for the purpose of evaluating  $O_3$  transport and recommending strategies for mitigating interstate pollution. The OTAG was a consultative process among 37 eastern states which included examination of the extent that  $NO_x$  emissions from hundreds of kilometers away are contributing to smog problems in downwind cities in the eastern half of the country, such as Atlanta, Boston, and Chicago. The OTAG completed its work in June 1997 and on July 8, 1997 forwarded its recommendations to EPA for achieving additional cost-effective emissions reduction programs to decrease ground-level  $O_3$  throughout the eastern United States. Based on these recommendations and additional information, EPA will complete a rulemaking action requiring States in the OTAG region that are significantly contributing to  $O_3$  nonattainment or interfering with maintenance of attainment in downwind States to revise their SIPs to include new rules to reduce their emissions of  $NO_x$ .

#### OTAG

- Reductions: Depends on EPA rulemaking and subsequent SIP revisions.

### Northeast Ozone Transport Commission (OTC) NO<sub>x</sub> Memorandum of Understanding (MOU) Phases II & III

To implement the OTC MOU, the required emissions reductions are applied to a 1990 baseline for NO<sub>x</sub> emissions in the OTR to create a "cap", or emissions budget for each of the two target years: 1999 and 2003. The NO<sub>x</sub> Budget Model Rule provides that once the 1990 baseline is established, the OTC MOU emissions reduction requirement is applied to create the 1999 and 2003 emissions budgets. The budget would then be allocated as "allowances" to the emission units subject to the program (budget sources). Budget sources are defined as fossil fuel fired boilers and indirect heat exchangers of 250 million Btu or greater, and electric generating units of 15 megawatts, or greater. Budget sources are defined on a unit level, meaning that each boiler or utility generator is considered a separate budget source. There are approximately 465 budget sources that would be applicable to the NO<sub>x</sub> Budget Program. The regionwide seasonal NO<sub>x</sub> Budget for 1999 (Phase II Target) is approximately 220,000 tons. For Phase III, in 2003, the target is approximately 143,000 tons of NO<sub>x</sub>.

#### OTC NO<sub>x</sub> MOU

Reductions: Approximately 272,000 ton seasonal (May through September reductions) reduction in Phase II (May 1999); and 76,000 additional ton seasonal reduction in Phase III (May 2003).
Reference: NESCAUM/MARAMA NO<sub>x</sub> Budget Model Rule, January 31, 1996 and 1990 OTC Nox Baseline Emission Inventory", Volume 1: Supplemental Material. E.H. Pechan Associates, July 12, 1995.

#### *Reasonably Available Control Technology (RACT)*

Certain existing major sources of  $NO_x$  must purchase and install reasonably available controls to decrease  $NO_x$  emissions. The new  $NO_x$  requirements apply in certain  $O_3$  nonattainment areas and in the Northeastern Ozone Transport Region.

#### RACT

- Reductions: generally a 30-50 percent reduction in NO<sub>x</sub> emissions.
- Reference: NO<sub>x</sub> Supplement to the General Preamble, November 25, 1992 (57 FR 55620).

#### Substitution of NO<sub>x</sub> for Required VOC Emissions Reductions

States may choose to decrease  $NO_x$  emissions instead of required VOC emissions reductions for certain VOC control programs.

#### Visibility & Regional Haze

The Administrator of EPA signed the notice of proposed rulemaking for the regional haze rules on July 18, 1997.

#### C. Sources and Sinks of Atmospheric Nitrogen

#### Sources of $NO_x$

#### Summary

A summary of national NO<sub>x</sub> emissions sources and NO<sub>x</sub> emissions trends is located in the "Introduction/Overview" section of this document. A map of large stationary sources (figure C-1) and an emissions density map (figure C-2) are provided (EPA, 1995). Supplemental information is provided below regarding sources of NO<sub>x</sub> emissions. In addition, information is provided regarding sinks of NO<sub>x</sub>.

#### Fuel Combustion: Electric Utility, Industrial, and Other

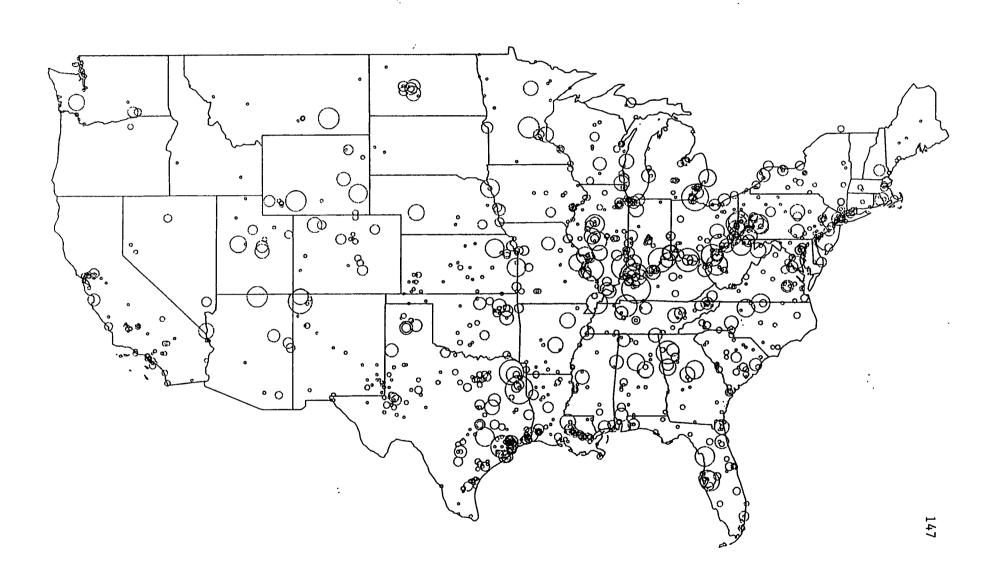
 $NO_x$  is emitted when fossil fuels are used to generate electricity. Electric utilities account for a large portion of the total national  $NO_x$  emissions; about 8 million tons in 1994, accounting for 33 percent of total  $NO_x$  emissions in that year as shown in tables C-1 and C-2 (EPA, 1995).

#### Transportation: On-Road Vehicles and Non-Road Sources

Emissions from on-road vehicles peaked in 1978 and have declined since then due to emissions control programs. Currently, on-road vehicle emissions constitute approximately 32 percent of total NO<sub>x</sub> emissions. Figure C-3 below displays trends in on-road NO<sub>x</sub> emissions, vehicle miles traveled, fuel use, and real gasoline prices for the period 1940 through 1993. NO<sub>x</sub> emissions from on-road vehicles increased as VMT and fuel use increased from the period 1940 through 1978 (EPA, 1995). However, NO<sub>x</sub> emissions begin to decline after 1978 while VMT and fuel use continued rising. The effects of regulations controlling vehicle emissions accounts for the declines in NO<sub>x</sub> emissions occurring after 1978. Although VMT has more than doubled since 1970, NO<sub>x</sub> emissions from on-road vehicles are nearly equal to their 1970 levels.

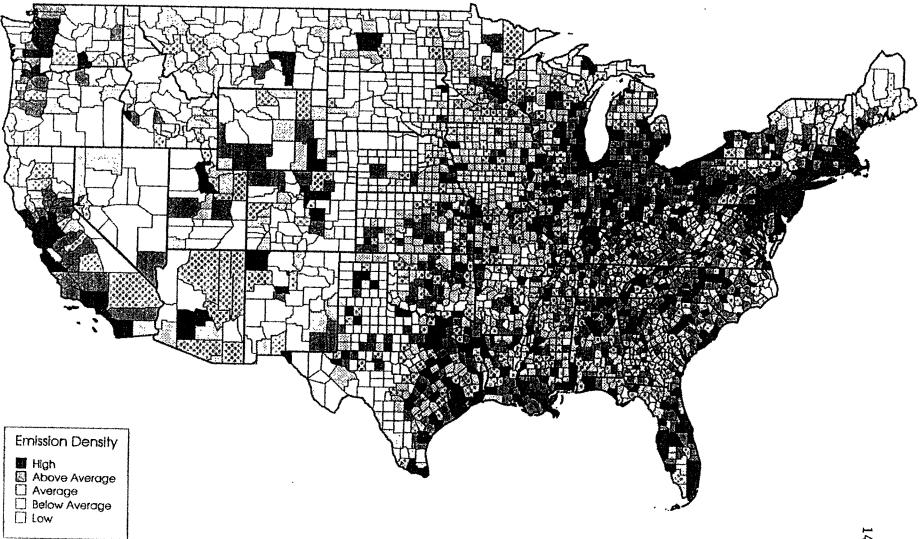
In contrast to the on-road vehicle  $NO_x$  emissions trends, emissions from non-road sources increased over the entire period of 1940 to 1994. Emissions control measures for selected non-road engine categories are scheduled to begin in 1996. Significant emissions reductions are not expected, however, until after the year 2000.

# Plants in 1990 with Greater than 1,000 tons per year of NITROGEN OXIDE Emissions



• ;

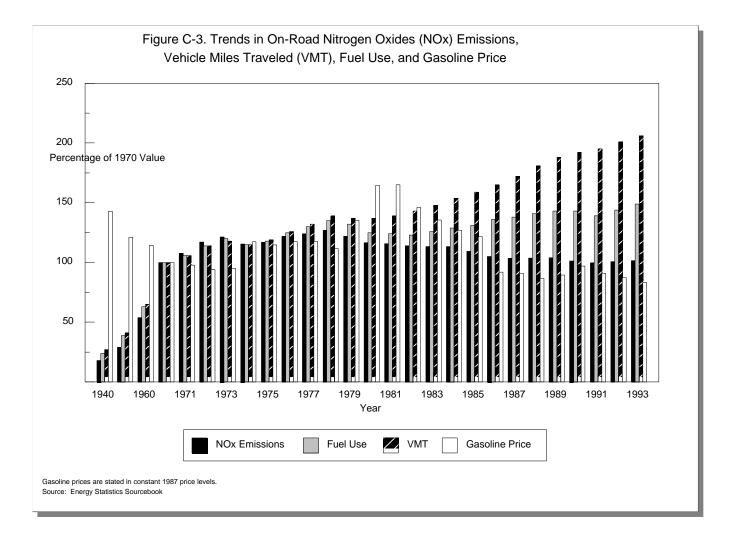
# **Density Map of 1994 NITROGEN OXIDE Emissions**



		NO <sub>x</sub>	
Source Category	Point	Area	Total
FUEL COMBELECTRIC UTILITY	7,795	0	7,795
FUEL COMBINDUSTRIAL	1,891	1,315	3,206
FUEL COMBOTHER	100	627	727
CHEMICAL & ALLIED PRODUCT MFG	291	0	291
METALS PROCESSING	84	0	84
PETROLEUM & RELATED INDUSTRIES	95	0	95
OTHER INDUSTRIAL PROCESSES	324	5	328
SOLVENT UTILIZATION	3	0	3
STORAGE & TRANSPORT	3	0	3
WASTE DISPOSAL & RECYCLING	20	65	85
ON-ROAD VEHICLES	0	7,530	7,530
NON-ROAD SOURCES	0	3,095	3,095
MISCELLANEOUS*	0	374	374
Total	10,604	13,011	23,615
Emissions (p	percent)		
		NO <sub>x</sub>	
Source Category	Point	Area	Total
	70 54	0.00	22.04

# Table C-1. 1994 National $NO_x$ Emissions by Source Category (thousand short tons)

		NO <sub>x</sub>			
Source Category	Point	Area	Total		
FUEL COMBELECTRIC UTILITY	73.51	0.00	33.01		
FUEL COMBINDUSTRIAL	17.83	10.11	13.58		
FUEL COMBOTHER	0.94	4.82	3.08		
CHEMICAL & ALLIED PRODUCT MFG	2.74	0.00	1.23		
METALS PROCESSING	0.80	0.00	0.36		
PETROLEUM & RELATED INDUSTRIES	0.89	0.00	0.40		
OTHER INDUSTRIAL PROCESSES	3.05	0.04	1.39		
SOLVENT UTILIZATION	0.02	0.00	0.01		
STORAGE & TRANSPORT	0.02	0.00	0.01		
WASTE DISPOSAL & RECYCLING	0.19	0.50	0.36		
ON-ROAD VEHICLES	0.00	57.87	31.88		
NON-ROAD SOURCES	0.00	23.79	13.10		
MISCELLANEOUS*	0.00	2.88	1.59		
Total	100	100	100		



#### 

Source Category	1940	1950	1960	1970	1980	1990	1993	1994
FUEL COMBELEC. UTIL.	660	1,316	2,536	4,900	7,024	7,516	7,773	7,795
Coal	467	1,118	2,038	3,888	6,123	6,698	7,008	7,007
bituminous	255	584	1,154	2,112	3,439	4,600	4,535	4,497
subbituminous	125	288	568	1,041	1,694	1,692	2,054	2,098
Oil	193	198	498	1,012	901	210	169	151
FUEL COMBINDUSTRIAL	2,543	3,192	4,075	4,325	3,555	3,256	3,197	3,206
Coal	2,012	1,076	782	771	444	613	550	568
bituminous	1,301	688	533	532	306	445	399	412
Gas	365	1,756	2,954	3,060	2,619	1,656	1,650	1,634
natural	337	1,692	2,846	3,053	2,469	1,436	1,440	1,427
FUEL COMBOTHER	529	647	760	836	741	712	726	727
Residential Other	177	227	362	439	356	352	363	364
CHEMICAL & ALLIED PRODUCT MFG	6	63	110	271	216	276	286	291
METALS PROCESSING	4	110	110	77	65	81	81	84
Ferrous Metals Processing	4	110	110	77	65	53	54	56
PETROLEUM & RELATED INDUSTRIES	105	110	220	240	72	100	95	95
OTHER INDUSTRIAL PROCESSES	107	93	131	187	205	306	315	328
Mineral Products	105	89	123	169	181	216	222	234
cement mfg	32	55	78	97	98	121	124	131
SOLVENT UTILIZATION	NA	NA	NA	NA	NA	2	3	3
STORAGE & TRANSPORT	NA	NA	NA	NA	NA	2	3	3
WASTE DISPOSAL & RECYCLING	110	215	331	440	111	82	84	85
ON-ROAD VEHICLES	1,330	2,143	3,982	7,390	8,621	7,488	7,510	7,530
Light-Duty Gas Vehicles & Motorcycles	970	1,415	2,607	4,158	4,421	3,437	3,680	3,750
light-duty gas vehicles	970	1,415	2,606	4,156	4,416	3,425	3,668	3,737
Light-Duty Gas Trucks	204	339	525	1,278	1,408	1,341	1,420	1,432
light-duty gas trucks 1	132	219	339	725	864	780	828	830
light-duty gas trucks 2	73	120	186	553	544	561	592	603
Heavy-Duty Gas Vehicles	155	296	363	278	300	335	315	333
Diesels	NA	93	487	1,676	2,493	2,375	2,094	2,015
heavy-duty diesel vehicles	NA	93	487	1,676	2,463	2,332	2,047	1,966
NON-ROAD SOURCES	991	1,538	1,443	1,628	2,423	2,843	2,985	3,095
Non-Road Gasoline	122	249	312	81	102	124	122	125
Non-Road Diesel	103	187	247	941	1,374	1,478	1,433	1,494
construction	70	158	157	599	854	944	1,007	1,076
industrial	NA	NA	40	75	99	125	131	136
farm	33	29	50	166	280	230	256	265
airport service	NA	NA	NA	78	113	144	152	159
Aircraft	0	2	4	72	106	139	147	153
Marine Vessels	109	108	108	40	110	173	183	188
Railroads	657	992	772	495	731	929	945	947
MISCELLANEOUS	990	665	441	330	248	373	219	374
TOTAL ALL SOURCES	7,374	10,093	14,140	20,625	23,281	23,038	23,276	23,615

### Table C-2. Total National Emissions of Nitrogen Oxides, 1940 through 1994<br/>(thousand short tons)

Categories displayed below Tier 1 do not sum to Tier 1 totals because they are intended to show major contributors. 1994 emissions estimates are preliminary and will be updated in the next report.

Tier 1 source categories and emissions are shaded.

#### Estimated Global Emissions of NO<sub>x</sub>

The tables provided earlier in this section describe national emissions of  $NO_x$ . The table below provides estimates of *global* emissions of  $NO_x$ , expressed in million metric tons of *N* (Schlesinger, 1992). The total estimate is 52.5 million metric tons of N.

Fossil Fuel Combustion	24
Soil Release	12
Biomass Burning	8
Lightning	5
Ammonia Oxidation	3
Aircraft	0.4
Transport from Stratosphere	0.1

Table C-3. Estimated Global Emissions of  $NO_x$  Typical of the Last Decade (Million Metric Tons N/year)

#### NO<sub>x</sub> Sinks: Removal Processes

The main mechanism that removes  $NO_x$  from the atmosphere is the conversion of NO and  $NO_2$  to nitric acid (HNO<sub>3</sub>) and the subsequent dry or wet deposition. The reaction of the OH radical with  $NO_2$  is the major gas-phase route to the formation of HNO<sub>3</sub> and it is the major homogeneous gas-phase sink for  $NO_x$  in the troposphere (NRC, 1991). HNO<sub>3</sub> is formed in the daytime reaction of  $NO_2$  with the hydroxyl radical (OH) and in nighttime reactions which form dinitrogen pentoxide which then hydrolyzes to produce HNO<sub>3</sub> (Science and Technical Support Work Group, 1997). Gaseous HNO<sub>3</sub>, formed from this reaction, undergoes wet and dry deposition, including, in some cases, combination with gaseous ammonia to form particulate phase ammonium nitrate. Dry deposition refers to the uptake of gases and particles at the earth's surface by vegetation, soil, and water. Wet deposition is the major loss route for atmospheric  $NO_x$  (Schneider et al, 1982). Wet deposition refers to the removal of gases and particles from the atmosphere through incorporation into rain, fog, and cloud water, followed by precipitation to the earth's surface. Additional radical termination reactions include reaction of  $NO_2$  with higher carbon number peroxy radicals to make organic nitrates and with peroxyacyl radicals to make PAN (Jeffries, 1995).

The oxidations of NO<sub>x</sub> leading to removal by HNO<sub>3</sub> or nitrates take place in hours to days, during which time the NO<sub>x</sub> compounds participate in a number of reactions in which they switch back and forth between the various intermediate NO<sub>x</sub> (Roberts, 1995; Jeffries, 1995). The lifetime of NO<sub>x</sub> (due to chemical conversion to HNO<sub>3</sub>) is estimated to be 1-2 days (EPA, 1993), while the residence time for nitrate is estimated to be 1-3 days (Schwartz, 1989). In the summer, the tropospheric lifetime of NO<sub>2</sub> (due to chemical conversion to HNO<sub>3</sub>) is estimated of the order of one day at mid-latitudes (Schneider et al, 1982).

#### Sources of Reduced N

As briefly noted in portions of this document, reduced N compounds--ammonia (NH<sub>3</sub>) and ammonium (NH<sub>4</sub><sup>+</sup>)--are also critical to many of the public health and environmental impacts associated with atmospheric N compounds. Emissions from fertilized agriculture and domestic animals account for over half the total estimated global NH<sub>3</sub> budget of the atmosphere reflecting the level of direct human impact in this area (Schlesinger and Hartley, 1992). The major sink for atmospheric NH<sub>3</sub> is conversion to NH<sub>4</sub><sup>+</sup>, which is deposited in dry particles or as a dissolved ion in precipitation.

#### Estimated Global Emissions of NH<sub>3</sub>

The table below provides estimates of *global* emissions of  $NH_3$ , expressed in million metric tons of *N* (Schlesinger and Hartley, 1992). The total estimate is 75.2 million metric tons of N per year.

Domestic Animals	32
Sea Surface	13
Undisturbed Soils	10
Fertilizers	9
Biomass Burning	5
Human Excrement	4
Coal Combustion	2
Automobiles	0.2

Table C-4. Estimated Global Emissions of NH<sub>3</sub> (Million Metric Tons N/year)

#### References

National Research Council, Committee on Tropospheric Ozone Formation and Measurement, *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, 1991.

Jeffries, H., "Photochemical Air Pollution" in *Composition, Chemistry and Climate of the Atmosphere*, edited by H. Singh, Van Nostrand Reinhold, NY, 1995, ISBN 0-442-01264-0.

Roberts, J.M., "Reactive Odd-Nitrogen (NO<sub>y</sub>) in the Atmosphere" in *Composition, Chemistry and Climate of the Atmosphere*, edited by H. Singh, Van Nostrand Reinhold, NY, 1995, ISBN 0-442-01264-0.

Schlesinger, W. H. and A. E. Hartley, "A Global Budget for Atmospheric NH<sub>3</sub>," *Biochemistry* 15: 191-211, 1992.

Schneider, T., and L. Grant, (Editors) *Air Pollution by Nitrogen Oxides*, 1982, Elsevier Scientific Publishing Company, Amsterdam, The Netherlands, p.252.

Schwartz, S.E., "Acid Deposition: unraveling a regional phenomenon," *Science*, 243:753-763, 1989.

U.S. Environmental Protection Agency, *Air Quality Criteria for Ozone and Related Photochemical Oxidants*, external review draft EPA/600/AP-93/004a, 1993.

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, *National Air Pollutant Emissions Trends*, 1900-1994, EPA-454/R-95-011, October 1995.

CAA:	Clean Air Act
$CO_2$ :	Carbon dioxide
HNO <sub>3</sub> :	Nitric acid
N:	Nitrogen
NAAQS:	National ambient air quality standards
NH <sub>3</sub> :	Ammonia
$\mathrm{NH}_{4}^{^{+}}$ :	Ammonium
$N_2O$ :	Nitrous oxide
NO:	Nitric oxide
$NO_2$ :	Nitrogen dioxide
$NO_x$ :	Sum of NO and NO <sub>2</sub>
<b>O</b> <sub>2</sub> :	Oxygen
O <sub>3</sub> :	Ozone
PPB:	Parts per billion
PPM:	Parts per million
PM:	Particulate matter; refers to a solid or liquid material that is suspended in the atmosphere.
PM <sub>2.5</sub> :	Particles with an aerodynamic diameter less than or equal to 2.5 microns
$PM_{10}^{-1}$ :	Particles with an aerodynamic diameter less than or equal to 10 microns
PSD:	Prevention of Significant Deterioration
SIPs:	State implementation plans
$SO_x$ :	Sulfur oxides
$SO_2$ :	Sulfur dioxide
VOCs:	Volatile organic compounds

D. Acronyms and Abbreviations

153