Irwin, J. 2005. *A Historical Look at the Development of Regulatory Air Quality Models for the United States Environmental Protection Agency*. Chapter 18 of *AIR QUALITY MODELING - Theories, Methodologies, Computational Techniques, and Available Databases and Software. Vol. II – Advanced Topics* (P. Zannetti, Editor). Published by The EnviroComp Institute (http://www.envirocomp.org/) and the Air & Waste Management Association (http://www.awma.org/).

Chapter 18

A Historical Look at the Development of Regulatory Air Quality Models for the United States Environmental Protection Agency

John S. Irwin

John S. Irwin and Associates[1]*, Raleigh, NC (USA)*
jsirwinetal@nc.rr.com

Abstract: Information about the development and use of regulatory air quality models, with an emphasis on those whose development was sponsored or promoted by the United States Environmental Protection Agency (EPA), is provided. A broad definition of regulatory is used here to include not only modeling used for setting specific emission limits, but also modeling used in developing EPA's agenda. The review outlines the major events in U.S. air quality legislation, noting the resulting influence on air quality model development. This partial review is meant to augment critical science reviews available elsewhere.

Key Words: air quality, Clean Air Act and its Amendments, model development.

1 Introduction

What caused the development of mathematical simulation models that describe the transport and fate of pollutants as they move through the atmosphere? Was this simply a consequence of intellectual curiosity? Was there a military need for characterization of poison gas dispersion, smoke obscuration effectiveness, or

[1] From 1975 to 2004, John Irwin was a NOAA employee, on assignment to the U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

radioactive fallout? Were such models spawned as a result of a fear of nuclear power generation facilities? Was it recognized as fundamental to developing objective strategies for the mitigation of hazards to human health from industrial emissions? If the answers to these questions were known, it would likely be attributable to all of the above concerns and to many not listed. Certainly the Atomic Energy Act in 1954 (which ultimately founded the Nuclear Regulatory Agency) and Public Law 159 in 1955 (which ultimately founded the Environmental Protection Agency) were instrumental in stimulating research activities. As fundamental as the causes for development, it is also important to recognize as with the development of all science understanding, many individuals contributed with partial solutions, which have been cobbled together to form what are called air quality simulation models. Even today, air quality models characterize the outward observable effects more so than the fundamental manner in which the processes happen. Thus, most of today's air quality models are rightfully characterized as first-order characterizations of the mean effects of transport, dispersion and fate. Even the emission characterizations are model estimates that are in many instances first-order approximations.

The current air quality model is really a system of models or sub-models. Each of the sub-models performs a function when needed. For buoyant emissions, the plume rise model attempts to estimate how high the mass of emissions will rise before stabilizing at some distance above the ground. A meteorological model (which in itself is a collection of models) characterizes the mean and turbulent properties of the atmosphere. A dispersion model estimates how the cloud of emissions expands as it moves downwind. A chemistry model simulates chemical transformations. There can be a wet deposition model to estimate the removal of mass by rainfall, and a dry deposition model to estimate the removal of mass to the ground and vegetation. As described by Peters et al. (1995), there is a jargon developing that is used to describe the level of sophistication in a modeling system. The first-generation models have first-order chemistry (with only a few primary reactions simulated). The transport and dispersion are founded on steady-state approximations in time and space. The second-generation models typically add removal processes, increase the level of sophistication in the parameterizations and chemistry simulations, and allow transport and dispersion to vary as a function of time and space. The third-generation models have yet to be realized, and currently are called the next-generation models. They will consist of select processes coupled together so that interactions and feedbacks can be investigated (e.g., aerosol formation attenuation of incoming radiation, which could alter aerosol formation rates).

There have been a number of reviews on the subject of the history of the development of air quality models. The five volumes edited by Stern (1976, 1977) cover the development of almost all of the aspects associated with air quality management, of which modeling is but a part. DeMarrais (1974), Turner (1979), and Randerson (1984) summarized the early years with emphasis on the characterization of the transport and fate of inert pollutants. Seinfeld (1988)

reviewed the early years in photochemical modeling. The transition from thinking that photochemical problems could be resolved with local-scale models to requiring regional-scale models can be seen in the reviews by Eliassen (1980), Fisher (1983), Eliassen et al. (1982), Peters et al. (1995), and Russell and Dennis (2000). The recent review by Seigneur et al. (1999) provides a review of particulate matter models.

There are several reviews that focus particularly on the Environmental Protection Agency (EPA), its history and the development of its programs. Jasanoff (1990) reviews several science policy disputes that involved EPA regulatory decisions, and the redress of increased use of science peer reviews and Science Advisory Boards. U.S. Environmental Protection Agency (1996) provides a summary of the first 25 years of EPA, its people and the legislative events of note. National Research Council (1991a) provides an excellent overview of EPA's attempts to devise legislative tools and modeling tools to assess ozone impacts. National Research Council (1991b) provides a review of how EPA envisioned an increased emphasis on reducing human health risks through exposure and how risk assessment would require revision of the modeling systems to address several new issues (e.g., the difference between ambient concentration values versus human exposures, the variability of human susceptibility to health risks, the increased uncertainties associated with such modeling systems). National Research Council (1994) provides a review of EPA's problems to successfully respond to legislative requirements to formally identify and then institute programs to reduce ambient concentrations of hazardous air pollutants.

The purpose of this discussion is to review the development of regulatory air quality models within the United States as viewed from the EPA perspective. EPA uses results from air quality modeling to define emission limits for new sources and for existing sources that are considering upgrades or changes to their process that would change their emissions. EPA also uses results from air quality modeling to investigate the consequences of alternative national pollution control strategies. In the discussion that follows, we use a broad definition of the term regulatory modeling to include both of these purposes. This review is intended to complement the cited critical science reviews. Since the inception of the EPA in 1970, the atmospheric scientists who provided EPA meteorological and air quality modeling support were with the Air Resources Laboratory (ARL) of the National Oceanic and Atmospheric Administration (NOAA)[2]. This resulted from a tradition within

[2] The Weather Bureaus Special Projects Research Field Office had three Field Research Divisions in 1948 to provide support to the emerging atomic energy program. Two of these Divisions served as a source of meteorological information and expertise for nuclear research facilities located near Oak Ridge, TN and Idaho Falls, ID. The Washington D.C. Division provided meteorological expertise regarding nuclear tests conducted around the world. In 1955, a division was established in Cincinnati, OH to provide meteorological consultation to the Public Health Service (PHS). In 1957, a Division was established in Las Vegas, NV to provide weather support for nuclear testing. In 1965, President Johnson consolidated two long-standing Department of Commerce agencies into the Environmental Sciences Services Administration (ESSA), the Coast and Geodetic Survey (established by President Jefferson in 1807) and the Weather Bureau (established by Congress in

the U.S. Weather Bureau to provide direct support to all Federal agencies needing meteorological information, which was passed on to the NOAA when it was formed. Most of the ARL scientists initially assigned to the EPA were former Weather Bureau scientists who had participated in many of the early investigations to construct operational transport and dispersion models. Therefore, it is easier to discuss the early years of air quality modeling if we start some years before the EPA was formally established.

Describing historical development poses a dilemma. For discussion purposes, it is easier if we focus on one topic; however, it creates a false sense of order, which is more related to the wisdom of hindsight. In reality, since everything is happening at once, it is difficult to determine the significance of individual developments when viewed in context. At various points, we will attempt to make some of the connections, but most of this work is left for the reader to assimilate. Section 2 of this discussion will focus on the legislative events that influenced and simulated the development of air quality models. Section 3 reviews the development of the early plume models for non-reactive pollutants, their evolution and specialization for characterizing dispersion from large individual industrial sources, and the current trend towards puff models. Section 4 summarizes the development of long-term air quality models that provide estimates of seasonal and annual average concentration for an urban area for non-reactive[3] pollutants. In the early to mid-1970s, these long-term models proved the feasibility of designing emission control strategies for entire cities for non-reactive pollutants, and thus offered a basis for considering development of air quality regulations. During the late 1990s as computers became more powerful, the use of long-term approximate solutions and long-term models seems to have declined. Section 5 reviews the development of tropospheric chemistry models, which were first designed solely for estimating ozone impact for cities. As experience and understanding were gained, it became clear that all secondary pollutants (e.g., ozone, sulfate, nitrates, etc.) involved regional-scale formation and transport. Section 6 attempts to summarize the issues (some resolved, many still pending) that are influencing current model development.

1891). In 1970, President Nixon combined ESSA with seven other earth science programs to establish the NOAA. By then, the ARL had five Divisions: Idaho Falls, Las Vegas, Oak Ridge, Washington D.C., and Research Triangle Park. The Cincinnati Office was moved to Research Triangle Park, NC in 1969 specifically to provide support to the EPA.

[3] In truth, all emissions undergo chemical transformations. The emphasis here is on those emitted directly into the atmosphere that undergo slow chemical transformations (if any).

2 Legislative History of Air Pollution Modeling

There are more than a dozen major statutes or laws that form the legal basis for the Environmental Protection Agency (EPA)[4] programs. There is a natural cause and effect linkage between legislation and air pollution model development, and we focus here on those that seem to directly affect air pollution model development. Air pollution models are used primarily for planning, as opposed to reacting to some emergency or accident. Albeit in an idealized sense, air pollution models provide a cost effective means for investigating the current and future possible conditions given certain assumptions. Given the expense of industry (and ultimately to the industry's customers) to install and maintain emission control equipment and procedures, air pollution models have been developed with new capacities as new requirements are either proposed for legislation or promulgated by legislation.

The Atomic Energy Act of 1954 required the Atomic Energy Commission (AEC), whose programs are now administered by the Nuclear Regulatory Commission (NRC) and the Department of Energy (DOE), to make a detailed environmental assessment before a nuclear construction permit and final operation license were issued. This was the first federal requirement for a systematic assessment of environmental impact that resulted in the routine use of mathematical simulation models for the characterization of the transport and fate of materials through the atmosphere.

Public Law 159, dated July 14, 1955, gave the Surgeon General of the Public Health Service (PHS), under the supervision of the Secretary of Health, Education, and Welfare (HEW), the responsibility of mitigating air pollution. Through an interagency agreement between the PHS and the National Weather Bureau, three meteorologists (Jack Lovett, Francis Pooler and Raymond Wanta) were assigned to support the PHS Air Pollution Engineering Center at the Taft Sanitary Engineering Center in Cincinnati, Ohio. Over the years, the Weather Bureau division in Cincinnati increased in number to 26 by 1967.

In 1963 the Clean Air Act (CAA) was passed. This act gave limited enforcement authority to the federal government, increased the availability of research and development money, and called for the development of air quality criteria (for when air is deemed adverse to public health or welfare).

The 1965 Amendments to the Clean Air Act provided federal authority to control emissions from new automobiles. The Air Quality Act of 1967 emphasized state control of air pollution problems and called for an expanded federal program. Friedlander and Seinfeld (1969) mentioned that the United States was being divided into about 100 air quality control regions. A major emphasis was to be

[4] (http://www.epa.gov/epahome/laws.htm) provides a list of these laws and links to the text of these laws and regulations.

placed on diffusion modeling in the delineation of the regions. Still there were no National Ambient Air Quality Standards (NAAQS)[5]. With no NAAQS and no time frames defined for attaining air quality standards, there was no national consistency in the approaches being considered.

In 1970, under Reorganization Plan Number 3[6], President Richard Nixon formed the U.S. Environmental Protection Agency using parts of HEW, the Department of Interior, U.S. Department of Agriculture, and the AEC. The Weather Bureau having earlier evolved into the Environmental Science Services Administration (ESSA) became the National Oceanic Atmospheric Administration (NOAA) under Reorganization Plan Number 4[7]. On December 31, 1970, the Clean Air Act was again amended. The amendments of 1970 revolutionized federal policies by establishing philosophies that dominate the EPA today; air quality was identified as a major public health problem, quantitative air quality management (e.g., modeling) was introduced, and the partnership between the federal and state agencies was clarified. The states were required to develop State Implementation Plans (SIP) to demonstrate, using air quality modeling, how they intended to attain and maintain the specified NAAQS within a specified time frame. NAAQS were instituted for six pollutants: carbon monoxide, total suspended particulates, sulfur dioxide, oxidants (revised to photochemical ozone and non-methane hydrocarbons in 1977), nitrogen dioxide and lead. Process-based models for ozone were yet to be developed, so an empirical method called Appendix J was used to estimate the percentage reduction (rollback) needed in total and non-methane hydrocarbon emissions to reduce maximum afternoon ozone concentration values. Congress set 1975 as the deadline for attaining these NAAQS. Section 112 of the 1970 Clean Air Act required EPA to set emission standards for hazardous air pollutants so as to protect public health with an ample margin of safety. As will be seen in the following discussion, EPA found control of ozone an elusive problem, and by 1984, EPA had listed only eight pollutants as hazardous[8].

By 1977, 2 years after the 1975 deadline specified in 1970, many areas were still in violation of the ozone NAAQS. The Clean Air Act Amendments of 1977 set a new compliance date of 1982 for attainment of the ozone and carbon monoxide

---

[5] National primary ambient standards are set to protect the public health and secondary standards are set to protect the public welfare. Each standard specifies an averaging time, frequency of occurrence and concentration value (e.g., a 1-hour concentration average not to be exceeded more than once per year). EPA is required by the 1970 Clean Air Act to review the primary and secondary standards at least once every five years to determine whether revisions to the standards are necessary to continue protecting public health and the environment. For more information, see http://www.epa.gov/ttn/oarpg/naaqsfin/naaqs.html.

[6] See http://www.epa.gov/history/publications/origins6.htm.

[7] See http://www.history.noaa.gov/eo11564.html and http://www.history.noaa.gov/index.html.

[8] The chemicals listed as hazardous air pollutants under the National Standards for Hazardous Air Pollutants (NESHAP), with month/year of public notice in parenthesis, were: asbestos (3/71), beryllium (3/71), mercury (3/71), vinyl chloride (12/75), benzene (6/77), radionuclides (12/79), inorganic arsenic (6/80), and coke-oven emissions (9/84).

NAAQS, and areas that demonstrated they could not meet the 1982 deadline were given extensions until 1987. The Clean Air Act Amendments of 1977 established the formal regulatory use of air quality dispersion models. These amendments established the concept of Prevention of Significant Deterioration (PSD) to be applied before construction of all new or modified major emitting facilities. Projected emissions from these facilities were to be modeled to determine if these sources (in combination with existing sources) would cause unacceptable ambient sulfur dioxide and particulate concentration values. Since PSD review is conducted before construction, the focus is on air quality modeling. Recognizing the burden placed on models by these legislative amendments, EPA was required to hold national modeling conferences at 3-year intervals to review modeling practices. EPA was further instructed to describe with reasonable particularity the modeling procedures and requirements. This led to the development of modeling guidance that described which air quality simulation model to use, how the meteorological conditions were to be processed for analysis, and the manner in which the modeling assessment was to be conducted.

Evidence was mounting by 1977 that ozone formation for the Eastern United States had a significant regional component that was beyond the reach of the Clean Air Act and its Amendments. Concerns were also being raised that regional transport might also be responsible for the observed elevated levels of sulfate. In 1980, President Carter signed the Acid Precipitation Act of 1980 (Public Law 96-294). The Acid Precipitation Act gave the government 10 years to perform a comprehensive assessment of the fate of sulfate. To address this requirement, the National Acid Precipitation Assessment Program (NAPAP) was established[9]. Looking back, 1980 can be viewed as a turning point for the EPA. From this point and on, the emphasis was to be on regional model development and evaluation issues.

By 1983, EPA had listed only six air pollutants as hazardous under the National Standards for Hazardous Air Pollutants (NESHAP), which was evidence of the difficulty of defining and implementing risk-based emission standards. In response to a directive from the Congress, the Food and Drug Administration (FDA), contracted with the National Academy of Sciences to examine whether alterations in institutional arrangements or procedures, particularly the organizational separation of risk assessment from regulatory decision-making and the use of uniform guidelines for inferring risk from available scientific information, could improve federal risk assessment activities. The Committee on the Institutional Means for Assessment of Risks to Public Health was formed in the National Research Council's Commission on Life Sciences in October 1981 and completed its work in January 1983 (National Research Council, 1983). As a general conclusion, the Committee believed that the basic problem in risk assessment was the sparseness and uncertainty of the scientific knowledge of the health hazards addressed, and this problem was seen as having no ready solution. These National

[9] See http://www.oar.noaa.gov/organization/napap.html.

Research Council recommendations planted the seed within the EPA culture to conduct risk assessments that include a formal uncertainty assessment on the methods and conclusions reached. A key goal of these risk assessments would be to maintain a clear separation between science and policy, and in which transparency (all assumptions explicitly stated) was a stated goal.

By 1990, 3 years after the extended deadline of 1987, more than 133 million Americans were living in the 96 areas that were not in attainment of the ozone NAAQS defined in 1977. The 1990 Clean Air Act Amendments (CAAA) required redefinition of ozone non-attainment into five classifications: extreme, severe, serious, moderate, or marginal. The classification and jurisdictional boundaries of the non-attainment areas required extensive analyses of monitoring and modeling results. Section 182 of the amendments stipulates for areas designated serious (16 in number), severe (8), or extreme (1), that within 4 years after the date of the enactment of the CAAA of 1990, the State shall submit a revision to the applicable implementation plan that includes "…a demonstration that the plan, as revised, will provide for attainment of the ozone national ambient air quality standard by the applicable date. This attainment demonstration must be based on photochemical grid modeling or any other analytical method determined by the Administrator … to be at least as effective". Section 184 of the 1990 CAAA established an interstate ozone transport region extending from Washington D.C. metropolitan area to Maine. The amendments also provided for creation of interstate transport commissions, to in effect, serve the role as broker between the several States included within an interstate ozone transport region, who were ultimately responsible for compliance with the provisions of the Clean Air Act and its Amendments.

In 1990, Congress revised the 1970 CAA procedures to be used to reduce health impacts from hazardous pollutants, by rewriting Section 112 in Title III of the 1990 Clean Air Act Amendments, and prescribing a two-stage program. Congress defined 189 compounds and compound classes as hazardous (subject to possible additions and deletions). In the first-stage of pollutant reduction, EPA was required to define technology standards and maximum available control technology (MACT) on sources of those chemicals. Section 112(f) defined the second-stage of the program, which required EPA to assess the residual risks associated with any emissions remaining, following installation of the MACT on the affected sources. A second-stage of controls is triggered only if EPA determines that an ample margin of safety has not been obtained by the technology-based emission reductions.

Title IV of the 1990 CAAA was the first national effort to use market-based incentives to achieve environmental goals, rather than the command-and-control approach employed to this point. To reduce the adverse effects of acid deposition, reductions in annual emissions of its precursors, and sulfur dioxide and oxides of nitrogen from combustion of fossil fuels, a program of alternative control measures was initiated that included technology adaptation (e.g., scrubbers and

higher-efficiency boilers), created a fuel switching, and emission allowance trading and banking system. With the use of market-based incentives to reduce adverse effects of acid deposition, the pressure to complete an operational acid deposition model was reduced, and the direction in model development turned away from sulfate acid deposition to developing characterizations of fine particles of all origins.

The Clean Air Act Amendments of 1990 defined new procedures for attaining pollution reductions for sulfur dioxide and toxics, which avoided direct use of modeling, and this had an impact on the development of air quality modeling. As will be discussed later, support for local-scale model development gradually diminished as more attention was given to development of large-scale grid models that could treat the chemistry of several pollutants simultaneously. The 1990 amendments required EPA to determine if local-scale plume models were systematically underestimating the impact of fugitive emissions (from roadways and dredging operations) associated with large surface coal mines. This provided a basis for minor improvements to be made in algorithms used in local-scale modeling for dry deposition. The regulatory importance of modeling was expanded from demonstrating compliance, as expressed in the amendments of 1977 and assessment of ozone abatement strategies, to being needed for assessment of the annual atmospheric deposition of toxic substances (sources and relative contributions) to the "Great Waters" (Great Lakes, Chesapeake Bay, Lake Champlain, coastal waters).

During the decade following the enactment of the CAAA of 1990, EPA completed reviews of the ozone and particulate NAAQS. In March of 1993, notice was given with a final decision not to revise the existing primary and secondary ozone NAAQS (58 FR 13008). Both were of the form of a 1-hour average not to be exceeded more than once per year, based on an analysis of 3 years of data. In July 1997, EPA made known its intention to revise the primary and secondary ozone NAAQS (Federal Register Vol. 62 No. 138) to the form of a 3-year average of the fourth highest maximum daily 8-hour average. In July 1997, EPA made known its intention to revise the primary and secondary particulate matter NAAQS. Lawsuits filed questioning whether the EPA had been given too much authority by the 1970 CAA were set aside by a Supreme Court Decision in February 2001[10]. The primary $PM_{2.5}$[11] particulate matter NAAQS was to have the form of a 3-year average of the 98th percentile of 24-hour PM2.5 concentration values. The primary PM10[10] particulate matter NAAQS was to have the form of a 3-year average of the 99th percentile of 24-hour PM10 concentration values. The secondary particulate matter NAAQS were revised to be identical to the proposed primary standards. Of interest to air quality modeling was that these proposed revisions to the ozone and particulate matter NAAQS continued the need for multi-year modeling

[10] See http://www.epa.gov/airlinks/airlinks4.html and http://www.epa.gov/ttn/oarpg/naaqsfin/.

[11] $PM_{2.5}$ refers to particulate matter with a diameter of 2.5 microns or less, and PM10 refers to particulate matter with a diameter of 10 microns or less.

assessments of the upper percentile values of the respective concentration distributions.

In summary, the legislation that has most stimulated the regulatory use of air quality modeling are: 1) Atomic Energy Act of 1954, as it established the requirement for Weather Bureau Field Offices to provide technical services in environmental impact assessment which ultimately established the linkage between NOAA and EPA; Public Law 159 in 1955, as it created the Public Heath Service that ultimately became the Environmental Protection Agency; 2) the Clean Air Act Amendments of 1970 and 1977, which relied on air quality modeling as a means for demonstrating compliance for the development of State Implementation Plans and for permits required by the New Source Review program; and finally, 3) the Clean Air Act Amendments of 1990, which formalized the requirement to use tropospheric chemistry air quality models for demonstrating a plan for attainment of the ozone NAAQS.

3 Air Quality Models for Individual Industrial Facilities

The PSD/NSR defined in the CAA amendments of 1977 established a need for air quality models that could be applied for major emitting facilities (to be constructed or to be modified) in order to determine if these sources (in combination with existing sources) would cause unacceptable ambient sulfur dioxide and particulate concentration values. As discussed initially in this section, the early models for sulfur dioxide and particulate emissions evolved from models constructed earlier for assessment of gas warfare and dispersion of nuclear fallout from atomic explosions. Based on predicted impacts from these dispersion models, the amounts of emissions allowed from each stack were estimated, such that the ambient air quality standards defined in the PSD/NSR program were attained. Before the PSD/NSR program, whether an area was in attainment was defined through air quality monitors. The PSD/NSR program allowed attainment to be defined through air quality modeling. In this section, we provide a brief review of the dispersion models used prior to 1977, and then follow the evolution of these models. It is sometimes distracting to follow a time sequence, and in these places we summarize the time sequence of the development of some major sub-model used in point source dispersion modeling.

In the early 1950s, there was great interest in nuclear fallout. Hyde (1952) used a trajectory analysis to show that radioactive debris from bomb tests in Nevada in October and November were detected in rainfall in France in November and December of 1951. List (1954) provided a detailed trajectory analysis for eight bomb tests conducted in the spring of 1952, including a discussion of fallout from three of the tests, with daily maps showing isolines of activity and areas of precipitation for several days following each of the tests. In discussing the possible relationships between detonations of atomic bombs and weather, Machta and Harris (1955) concluded that it was unlikely that such explosions affected the

weather. Both Bob List and Lester Machta were meteorologists assigned to the Washington D.C. Weather Bureau Field Division.

In 1955, the first addition of *Meteorology and Atomic Energy* (U.S. Weather Bureau, 1955) was published. The editor of this work was Harry Wexler who was then the Chief of the Scientific Services Division in the Weather Bureau. Personnel from the Scientific Services Division assisted in the collection and preparation of this report, many of which became commonplace names within air pollution research (e.g., F. A. Gifford Jr., B. List[12], L. Machta[6], D. Pack, F. Pooler). Written for the characterization of the transport and dispersion of nuclear radiation, this was the first compendium dealing with all the facets of air pollution meteorology. It included information on sources, development of dispersion patterns, methods and nomograms for computing downwind concentrations, meteorological instruments and their proper use and climatological data pertinent to air pollution meteorology.

At this point in time, the practical method for characterizing dispersion from a point source followed the ideas of Sutton (1947)[13]. During the summer of 1956, an experimental program to study micrometeorology and dispersion from near-surface releases was conducted near the town of O'Neil in north central Nebraska. This comprehensive turbulence and diffusion program of 70 tracer experiments was given the name Project Prairie Grass, and was instrumental in providing a basis for the development of practical methods for the characterization of atmospheric dispersion. One of the first papers to discuss these data was by Cramer (1957), in which a Gaussian plume model was described that related the horizontal and vertical dispersion to the observed standard deviations of lateral and elevation angles of the wind fluctuations. Barad and Haugen (1959) used the Project Prairie Grass results to specifically investigate the veracity of Sutton's model for plume dispersion. Not long thereafter, Pasquill (1961) offered a pragmatic technique to estimate Gaussian plume vertical and lateral dispersion that could be implemented with easily acquired meteorological observations, namely: insolation and wind speed. By 1958, F. Pasquill was corresponding with F.A. Gifford (who was then with the Oak Ridge Field Research Division), and R. McCormick and D.B. Turner (who were then with the Cincinnati Field Research Division) prior to 1961. So, Gifford (1961) offered a conversion of Pasquill's angular spread values to standard deviations of plume spread, and Turner (1961) offered a conversion of Pasquill's

---

[12] In 1949, Bob List edited sixth edition of the Smithsonian Meteorological Tables, which is now in its fifth reprint (List, 1971), a fundamental meteorological handbook. Lester Machta was hired at the inception of the Weather Bureau Field Offices in 1948, and was Director of the NOAA Air Resources Laboratories from 1968 to 1991, renamed in 1982 to NOAA Air Resources Laboratory.

[13] "… The incidence of gas warfare in 1916 gave an impetus to the investigation of atmospheric diffusion, which ultimately led to the formation of a special meteorological research team at the Chemical Defense Experimental Station, Porton, Wiltshire. The present paper is an integrated account of certain mathematical investigations carried out by the writer between 1932 and 1938. Some parts of this work were published in 1932 and 1934, but hitherto no connected account has existed outside of official reports."

stability classification criteria that employed hourly airport observations. These extensions to Pasquill's scheme simplified its use, and made it practical for it to be converted to a numerical algorithm.

Development of an air quality simulation model for use in air quality planning, involved the combination of two important concepts besides the characterization of plume dispersion. Industrial stacks emit heated gases that tend to rise, which requires a plume rise model. Elevated temperature inversions were known to contribute to the buildup of smoke and air pollution (smog) (Schrenk et al., 1949; Wilkins, 1953; Lucas, 1958), which requires a mixing height model. Characterizing the emission and dispersion of pollutants in an entire city involves treating hundreds of individual stack emissions in conjunction with low-level diffuse emissions. An early proof of concept was Turner's (1964) simulation of sulfur dioxide emissions for Nashville, TN, which illustrated that it was feasible to combine the concepts of Pasquill plume dispersion with the Holland (1953) plume rise and mixing heights using Holzworth's (1964) concepts, into a practical air quality simulation model. These results were based on a one-year study of Nashville, TN, where data from a network of 32 sampling stations were used. Results showed that the 24-hour sulfur dioxide concentrations and the aerial extent of the pollution impacts from multiple sources could be analyzed using a source inventory coupled with a numerical dispersion model. Turner (1967) explained the details of this type of model in a handbook with example problems and solutions for teaching the concepts.

The development of mixing height models is hampered by the fact that the depth through which emissions are mixed is an asymptotic result, that is best observed by analysis of vertical profiles of inert tracers released specifically for the purpose. Holzworth (1964) published a climatology of urban morning and afternoon mixing depths for the contiguous United States[14]. This report popularized the idea of using easy to determine mixing depths in air pollution evaluations. This report also illustrated the usefulness of numerical methods for the processing of large quantities of data for the practical evaluation of air pollution problems. Mixing depth data is rarely a primary focus of field studies; and thus, there are few quality data sets for use. Since Holzworth's investigations, EPA has not invested resources toward developing improved mixing height algorithms for use in its air quality models, but has chosen to select algorithms developed by others. As discussed in a recent review, Seibert et al. (2000), all available methods have strengths and weaknesses.

The early plume rise formulas were based on an integral model, in which the differential equations governing the total fluxes of mass, momentum and energy

[14] The morning urban mixing depth was determined as the intersection of a morning urban minimum potential temperature (the observed minimum temperature plus 5^{o} C) with the morning potential temperature profile (defined from a morning upper-air observation). The afternoon maximum mixing depth was determined as the intersection of the afternoon maximum potential temperature with the morning potential temperature profile.

through a plume cross section are closed using an entrainment assumption. Typically, the entrainment assumption[15] specifies that the average rate at which outside air enters the plume surface is proportional to the characteristic vertical velocity of the plume at any given height, and assumes that dilution by atmospheric turbulence is negligible in comparison to other factors (Taylor, 1945). Holland (1953), in describing the micrometeorology associated with the Oak Ridge, TN area, provided a plume rise formula that became known as the Holland formula. Scorer (1959) presented formulas for computing plume rise, claiming the existing formulas were too complicated for practical use. Then Briggs (1965), using dimensional arguments introduced a new plume rise for buoyant plumes. Nonhebel (1965) reviewed many plume rise formulae and noted the confusion. By 1969, there were 30 plume rise models to choose from, so the critical review by Briggs (1969), which offered a simplified alternative, was well received. G.A. Briggs extended his works over the time period from 1969 to 1975, which by then had become the primary plume rise model of choice (Briggs, 1975). As discussed by Weil (1988) and Netterville (1990), the plume rise model of Briggs was chosen to embody the primary physical processes of buoyant plume rise, when the ambient turbulence is small, and thus the dilution of the buoyant plume is dominated by its own motion, which is an apt characterization of plume rise in thermally neutral and stable atmospheric conditions. There are other conditions when these assumptions prove to be inadequate; namely, the penetration of buoyant fluid into an elevated inversion (where the thickness and strength of the elevated inversion complicates the characterization of the plume rise), and buoyant rise in a strongly convective boundary layer (where the strength of the convective eddies can exceed the buoyancy forces within the plume). To date, the models used routinely in air pollution assessments have continued to employ Briggs' plume rise model, and to treat the effects of penetration and rise in a convective boundary layer as special cases. This is in contrast to adopting plume rise models with a broader range of applicability. In part, this relates to the fact that, regardless of the plume rise model employed, the estimated rise characterizes the ensemble average plume rise, and there is little skill perceived (more variance explained) through the use of the more comprehensive plume rise models.

Soon after the move of the Cincinnati office to Research Triangle Park, NC in 1969, W. Snyder joined the NOAA staff on assignment to support the EPA. W. Snyder was a specialist in the operation and use of wind tunnels. Since there was no wind tunnel when he first arrived, his early years were devoted to constructing a wind tunnel, and establishing criteria for its use in simulating atmospheric turbulence and dispersion. Snyder (1972) laid the groundwork by defining the similarity criteria for modeling atmospheric flows in air and water using wind tunnels and water towing tanks.

---

[15] A reasonable assumption, since the focus was on characterizing the rise of a heated plume resulting from an atomic bomb blast.

One of the first questions posed to the EPA Wind Tunnel and Fluid Modeling Facility was whether flow around scale models of buildings could be used to investigate the potential for stack gas emissions to be captured in the lee wake of the buildings. Using results from wind tunnel simulations, Snyder and Lawson (1976) showed that for stacks close to buildings, designing a stack to be 2.5 times the building height was adequate for a building whose width perpendicular to the wind direction was twice its heights, but defined stack heights greater than needed for tall thin buildings. These investigations were extended by Huber and Snyder (1976) towards development of algorithms that could be incorporated into plume dispersion models to simulate the enhanced dispersion of plumes caught in building wakes.

By the early 1970s, D.B. Turner had a collection of numerical algorithms that were written in FORTRAN, and provided a means for simulating plume dispersion. Using an existing nationwide computer network, UNAMAP (User's Network for Applied Modeling of Air Pollution) came into being in 1972 (Turner et al., 1989), and consisted of six air quality simulation models. The computer network was only available to a limited few who had access, so it was decided to distribute the models as FORTRAN code on a magnetic tape, with the National Technical Information Service (NTIS) as the provider of the tape to the public. In 1989, the EPA Office of Air Quality Planning and Standards took over the responsibility of distributing the models to the user community using a dial-up Bulletin Board Service, which evolved to what is called today the Support Center for Regulatory Air Models (http://www.epa.gov/scram001/).

By 1977, the primary features of the models being used for characterizing dispersion from isolated point sources was: the vertical and lateral dispersion were characterized using the Pasquill-Gifford dispersion curves of 1961 for rural environs, the six[16] stability categories ranging from very unstable (A) to very stable (F) were characterized using D.B. Turner's 1961 criteria, the plume rise was characterized using the Briggs 1975 modeling equations, and the mixing height was characterized using the Holzworth 1964 model. For urban environs, the stability categories at night are forced to neutral (D). The model for modeling individual point sources was CRSTER (U.S. Environmental Protection Agency, 1977)[17]. A Gaussian plume for multiple sources, called RAM, was released in 1978 (Turner and Novak, 1978; U.S. Environmental Protection Agency, 1978a)[18].

---

[16] Pasquill (1961) had seven stability categories, and had envisioned the neutral category as being composed of two cases, adiabatic for when the surface is actively heated (daytime) and subadiabatic for when the surface is not heated (overcast and nighttime). EPA chose to use the subadiabatic category and its dispersion characterization for all neutral cases. This may explain, in part, a tendency for the resulting models to underestimate surface concentrations from tall stacks during daytime neutral conditions.

[17] EPA desired to quickly develop the FORTRAN code for modeling a single point source, and it became known as the crash (CRS) program for its tight time deadlines. When the scope was expanded to include terrain interactions, the model name was coined, CRSTER.

[18] Named after Robert A. McCormick who was the Director of the NOAA Meteorological Sciences

RAM was one of the first operational models used by the EPA to employ the St. Louis dispersion curves (McElroy and Pooler, 1968)[19] for characterizing urban dispersion effects. Using EPA wind tunnel research results for modeling building wake effects, CRSTER was adapted to become the first version of the Industrial Source Complex (ISC) model (U.S. Environmental Protection Agency, 1979).

Responding to the Clean Air Act Amendments of 1977, the *Guideline on Air Quality Models* (U.S. Environmental Protection Agency, 1978b) was published, which was to be used by EPA, States, and industry to prepare and review PSD air quality modeling assessments and State implementation plans. The Guideline of 1978 was intended to ensure consistent air quality analyses for modeling activities. This Guideline was developed and published for the specific purpose of complying with the 1977 Clean Air Act amendments that required EPA to prescribe with "reasonable particularity" the modeling procedures to be used in PSD assessments. The Guideline of 1978, in order to be reasonably prescriptive, had identified for each perceived situation, the model that would be recommended for regulatory assessments. With updates to the Guideline new situations were added. Example situations included: relatively flat terrain with rural or urban conditions, complex terrain with rural or urban conditions, long range transport, dispersion in the vicinity of the shoreline or ocean. The Guideline further stipulated the procedures an applicant would have to follow in order to employ an alternative model to that which was specified for use in the Guideline. Basically, the model had to be demonstrated to perform as well or better than the model prescribed in the Guideline for the particular situation. This proved to be an almost insurmountable requirement, except in a few circumstances where the applicant had sufficient resources and perseverance.

Around this time, EPA was sued by private industry for requiring the use of the Pasquill stability, a dispersion curves for the characterization of dispersion from isolated industrial sources having tall stacks. From 1976 through 1980, this lawsuit made its way through the court proceedings. The court ruled in favor of the EPA's position (FR, 1980). This ruling did not review the technical decision to use the Pasquill-Gifford dispersion curves for elevated releases (and in particular the dispersion curve associated with very unstable conditions, stability category A). What the courts did in coming to this decision was scrutinize EPA’s decision making process to insure that EPA’s decisions had not been arbitrary or capricious. The courts upheld EPA’s decision making process.

The publication of the Guideline in 1978 had several consequences. It recommended specific models for use in complying with the modeling required by the EPA for the Prevention of Significant Deterioration (PSD) and New Source Review (NSR) programs. It defined the EPA Regional Offices as having the final

---

Modeling Division when it was established in Research Triangle Park, NC in 1970.

[19] A reanalysis by G.A. Briggs of the St. Louis dispersion curves resulted in the Briggs urban curves, which were used in subsequent releases of EPA’s dispersion models (Gifford, 1976).

authority for interacting with the public, and reviewing and accepting PSD/NSR modeling results. It defined the process by which new models would be considered, and process by which the Guideline would be periodically updated. Through the years, the system defined in the 1978 Guideline for managing the PSD/NSR modeling requirements has proven itself to be highly effective and workable, both for regulators and those regulated[20]. There were two negative consequences resulting from the 1978 Guideline. First, the procedures for using alternative models required one to determine that the alternative model being proposed would perform better than that recommended in the Guideline. In his critical review of dispersion modeling, Turner (1979) noted that there were no recognized model performance standards (metersticks)[21]. With no accepted measure for defining a difference in performance, arguing the case for use of an alternative model was difficult. Secondly, in order to introduce a new model or replace an existing Guideline model, EPA had to follow the administrative requirements for publication of a proposed revision, a public review and comment time period, and then publication of the final revisions to the Guideline. This imposes an inertia in the revision of the modeling guidance that is at least 18 months or more in length. Even though results from research studies during the period from 1975 to 1995 proved to be one of the more exciting, providing many improved methods for the characterization point source dispersion, the structure and form of the models used for regulatory assessments remained largely unchanged.

Section 123 of the CAA Amendments of 1977 states that construction of tall stacks for the principal purpose of reducing pollutant impacts at the surface was not acceptable. It was considered good-engineering practice (GEP) to build stacks to avoid being captured within the wake effects of nearby buildings and obstacles (terrain effects). Guidance was published by the EPA on how to use a wind tunnel to determine the stack height needed to avoid building effects (Lawson and Snyder, 1983) and terrain effects (U.S. Environmental Protection Agency, 1985a) that enhance dispersion (result in concentration maxima that are 40% higher than would otherwise occur if the building or terrain were not affecting the flow).

---

[20] Since its inception, J.A. Tikvart of the US EPA has provided oversight and management of all activities associated with the drafting, updating and implementation of the Guideline. He foresaw that the purpose of the Guideline would be realized by the people who implemented the guidance. Through his efforts, various programs and activities were established, as the EPA Model Clearinghouse to resolve problematic technical questions, and annual workshops were held to ensure communication between the EPA Regional modeling contacts.

[21] J.A. Tikvart of the US EPA and L. Niemeyer of the ASMD established a Cooperative Agreement with the American Meteorological Society in 1979 to provide expertise and assistance in evaluating technical aspects of air quality models that may be used for applications as required by the Clean Air Act Amendments of 1977. The AMS Steering Committee for the EPA Cooperative Agreement fostered improved model evaluation metersticks through workshops and reviews (Randerson, 1979; Fox, 1981; Fox, 1984; Smith, 1984; Irwin and Smith, 1984). This committee also attempted to foster development of improved methods for characterizing transport and diffusion, which culminated in the formation of the AMS EPA Regulatory Model Improvement Committee (AERMIC), which developed AERMOD (Weil, 1992).

Another area where the EPA Wind Tunnel and Fluid Modeling Facility had a major impact was in developing a pragmatic method for characterizing the flow and dispersion around an isolated hill. Through a combination of wind tunnel and fluid towing tank experiments, Hunt and Snyder (1980) developed the dividing streamline concept, which suggests that unless the flow has sufficient energy, it will flow around an obstacle rather than flow directly over the obstacle. Full-scale tracer field studies confirmed these findings at Cinder Cone Butte, ID in 1980; Hogback Ridge, NM in 1982, and the Tracy Power Plant, NV in 1984 (Snyder et al., 1985). From these results, the EPA complex terrain dispersion model, CTDM/PLUS, was developed (Perry, 1992a, b).

During the period from 1975 to 1990, a new understanding was reached concerning dispersion within the convective boundary layer (CBL). The convective tank experiments by Willis and Deardorff (1976, 1978, 1981) revealed that the surface releases tended to slide along the surface until captured within a convective updraft. A surface release thus captured would appear to have an accelerated vertical dispersion in comparison to its lateral dispersion, as suggested by the Pasquill stability category A curve for vertical dispersion during very unstable conditions. An elevated release tended to slowly descend towards the surface, being caught within the general downdraft area surrounding the isolated convective updrafts. This view argued for a more complex dispersion model than was possible by the simple Gaussian plume model. These fluid modeling results were confirmed by the Convective Diffusion Observed by Remote Sensors (CONDORS) field experiments conducted at the NOAA facilities in Boulder, CO during August and September 1983 (Eberhard et al., 1988; Briggs, 1989, 1993a, b). Operational plume dispersion models were developed to address this new understanding of convective dispersion by the Maryland Power Plant Research Program (Weil and Brower, 1984; Weil and Corio, 1988); the EPA (Turner et al., 1986; Gryning et al., 1987), and EPRI (Hanna and Paine, 1989; Hanna and Chang, 1991). The most recent proposals have been ADMS (Carruthers et al., 1994, Bennet and Hunter, 1997, Owen et al., 2000) and AERMOD (Weil, 1992; Perry et al., 1994; Lee et al., 1996).

EPA has always had models to propose for use for short-range dispersion, but there has been a continuing need for models that could handle the more complex effects associated with dispersion involving transport beyond the near-field, say 15 to 20 km. Starting in 1975, EPA has supported the development of a series of models starting with MESOPUFF (Benkley and Bass, 1979), which was extended by the North Dakota State Department of Health to become MSPUFF (Schock and Weber, 1984). Petersen (1982) provided a model for handling an instantaneous release as a puff, which he then extended to handle a time-varying emission rate (possibly moving source, like a ship in a harbor) by a multi-source Gaussian puff model called INPUFF (Petersen and Lavdas, 1986). EPA supported the development of a multi-layer model, MESOPUFF II (Scire et al., 1984), that could better address wind shear effects on dispersion (variation of wind direction with height). MESOPUFF II was revised to become CALPUFF (Scire et al., 2000a, 2000b),

which has since received further development support (e.g., State of Victoria, Australia funded development of Graphical User Interfaces; U.S. National Forest Service inclusions of algorithms for treatment of forest fires; EPA funded aqueous phase chemistry and near-field dispersion enhancements; Aluminum Industry funded inclusion of buoyant line source algorithms; California energy commission funded inclusion of convective dispersion algorithms). Puff models can directly address the inhomogeneity in the meteorological conditions, which is impossible to address in the context of a steady-state plume dispersion model. Developing the terrain induced flows, storing the time-varying three-dimensional meteorological fields, and tracking possibly tens of thousand puffs have restricted puff models to large mainframe computers until recent times.

In 1999, EPA proposed that the ISC plume dispersion model be replaced with the second-generation plume dispersion model, AERMOD. It was also proposed that the Gaussian puff model, CALPUFF, be accepted for all refined modeling involving transport with distances greater than 50 km, and on a case-by-case basis for any situation involving complex winds (e.g., calms and stagnation, narrow valley channeling, dispersion near shorelines of large lakes and oceans). Application of either of these models anticipates a more sophisticated user than that envisioned in the development of the ISC modeling systems. As experience is gained in the use of the CALPUFF modeling system, it is anticipated that the realism provided by treating the time-variations of the three-dimensional wind and turbulence fields along with transformation and deposition will become highly desired, such that plume models may fall into disuse. For this to occur, various enhancements will be necessary in order to allow ready application of this modeling system to the variety of situations now handled by plume dispersion models.

There are several scientists whose names are distinguished in the development and use of models for simulating the impacts of individual industrial facilities in regulatory assessments. F. Pasquill and F.A. Gifford offered a pragmatic update of O.G. Sutton's model for characterizing the vertical and lateral extent of stack emissions as they disperse downwind. Through the publication of a practical handbook (a conversion of the subjective criteria for defining Pasquill's stability categories into objective criteria capable of being implemented in computer software, and establishment of a system for distribution of the numerical dispersion models), D.B. Turner popularized the use and application of Gaussian plume models in regulatory assessments. G.A. Briggs offered a pragmatic description of buoyant plume rise that could be easily used with limited definition of the meteorological conditions. W. Snyder's wind tunnel studies provided a basis for developing models of how the swirl of eddies around buildings and hills affect the plume rise and the dispersion of buoyant stack emissions. The efforts of these scientists formed the basis and use of industrial source dispersion models in regulatory assessments, not only within the United States, but also in other countries.

4 The Development of Urban-Scale Long-Term Air Quality Models

The Clean Air Act is a federal law covering the entire country, with the states doing much of the work. Under this law, EPA sets limits on how much of a pollutant can be in the air anywhere in the United States. This ensures that all Americans have the same basic health and environmental protections. The law allows individual states to have stronger pollution controls; but, states are not allowed to have weaker pollution controls than those set for the whole country. The law recognizes that it makes sense for states to take the lead in carrying out the Clean Air Act, because pollution control problems often require special understanding of local industries, geography, housing patterns, etc. States have to develop state implementation plans (SIP) that explain how each state will ensure that the limits set by the EPA will be reached or maintained. These plans invariably involved the use of air quality models to relate the control of emissions with estimated air quality impacts.

In the early years of air dispersion modeling (say, prior to 1968), most calculations were completed with paper, pencil and hand calculators. Early computers were limited in their memory capabilities. This spawned the development of a particular type of dispersion model, which employed a statistical summary of meteorological conditions, which then required a special algorithm for characterizing the resulting dispersion. These models came to be known as long-term models, as they were designed to provide annual or seasonal-average concentration values. These models have historical importance as they provided the early demonstrations of how air pollution dispersion models could be used to design emission control programs. They came into being around 1965 and survived for about 30 years, until computer memory and speed made them unnecessary.

Early examples of this type of model were described by Meade and Pasquill (1958) and Lucas (1958). The idea was relatively simple, but most of the algorithms for characterizing the basic processes (e.g., buoyant plume rise, plume dispersion, depletion, etc.) were simplistic (first-generation) with little experimental verification. Basically, a computation was made for each expected wind speed and stability condition, whose probability of occurrence was computed for wind sectors surrounding the source (varying from 12 to 16 wind sectors). The average concentration was computed by summing for each wind sector, the computed concentration at each downwind distance, multiplied by the frequency of occurrence of each wind speed and stability combination. Over the next 20 years, a series of climatological or long-term models were developed and tested that were based on this algorithm.

Pooler (1961) used the long-term algorithm, and was one of the first investigators to employ numerical methods for automating the computations (IBM 650 computer) to provide estimates of monthly average concentration values for

comparison with observations of sulphur dioxide (SO_2) collected daily from November 1958 through March 1959 at 123 sampling sites in Nashville, TN. We have to temper Pooler's evaluation results, as regressions were performed with the observed concentration values to provide best estimates of the variation of the monthly emission rates from the known sources. That said, the model overestimated[22] the observed values by a factor of 1.37 with 110 of the 122 values within a factor of two of the observed values, with 74 of the 122 values within 30%.

Gifford and Hanna (1970) offered an alternative to employing Gaussian plume modeling for simulating pollutant impacts from area source emissions. They demonstrated, through comparisons with other algorithms, that one can rely on compensation from adjacent area sources such that the lateral dispersion can be neglected. This was called the narrow-plume hypothesis by Calder (1969, 1977), and relies on the area source emissions to be something like a checker-board, with similar emissions in adjacent grid squares. Gifford and Hanna (1973) extended their discussion and demonstrated that annual or seasonal average concentrations might be approximated for an entire urban area using a simple relationship of $C = kQ/U$, where C is the average concentration (grams per cubic meter), k is the proportionality constant, Q is the average emission rate of the pollutant (grams per second per square meter, estimated as the total emissions for the city divided by the area of the city, which in their studies was typically of order 7 to 18 km in radius), and U is the annual or seasonal average wind speed (meters per second). The proportionality constant was determined to be mostly a function of stability (day versus night, etc.), and to a lesser extent city size (increasing as city size increased).

For the purposes of estimating an annual average concentration, it was found that k should be specified based on whether the emissions being characterized are elevated or near-surface, and for near-surface emissions, whether the receptors are very near or somewhat away. For near-surface releases, k is equal to 600 for receptors located very near the emissions (like roadway emissions of carbon monoxide), and k is equal to 250 for receptors located away from the emissions. For elevated releases, k is equal to 30 (Gifford and Hanna, 1973; Hanna et al., 1982).

Martin (1971) continued the development of the model used by Pooler with a comparison of results computed (IBM 1130) for a winter season of the average sulfur dioxide concentration values for comparisons with observations collected daily from December 1964 through February 1965 at 40 sites in the St. Louis area. Uncertainties in locating several large point sources precluded the use of results at 5 sites. Model estimates at 34 of the 35 remaining sites were within a factor of 2, with 14 within 30%. A reanalysis of the same data was performed by Calder (1971) using the Climatological Dispersion (CDM) model, and using a revised

[22] Reported factor of over or under estimation and correlation coefficient (r^2) was deduced through a linear regression with the intercept forced to be at the origin.

characterization of the area source emissions by Turner and Edmisten (1968). A major enhancement within the CDM over the model employed by Martin was to include an area source algorithm based on the narrow plume hypothesis (Calder, 1977). In spite of the attempts to improve the characterization of area source emissions and the dispersion from these low-level sources, the comparison results were similar to those achieved by Martin. Calder speculated that possible factors contributing to the tendency to overestimate the observed concentration values were: an inherently crude emissions inventory, no day versus night variation in emission rates, and the crude estimates of mixing height employed.

Turner et al. (1971) summarized the results obtained in applying the CDM model to estimate the annual average particulate and Sulfur dioxide concentration values for the New York area for 1969. Sulfur dioxide observations were available for comparison at 75 locations, and total suspended particulate matter observations were available for comparison at 113 locations. This version of the CDM employed the Briggs (1969) plume rise algorithms (in contrast to use of the Holland (1953) algorithms used by Martin and Calder in the St. Louis comparisons). For sulfur dioxide, it appears the CDM tended to slightly overpredict[2] concentration values by a factor of 1.11. Seventy-one of the 75 values were within a factor of 2, with 47 values within 30%. For particulates, it appears the CDM tended to slightly underpredict[2] concentration values by a factor of 0.93. 111 of the 113 values were within a factor of 2, with 94 within 30%.

Irwin and Brown (1985) applied the CDM model to estimate 1976 annual average sulfur dioxide concentration values for the St. Louis area. There were 13 sites, but omission of a lead smelter from the emission inventory precluded use of data at two sites for model performance comparisons. The emission inventory and monitoring results were obtained as part of the St. Louis Regional Air Pollution Study (Strothmann and Schiermeier, 1979). These simulations differ with those computed by Turner et al. in that urban dispersion parameters were used, based on tracer studies conducted in St. Louis (McElroy and Pooler, 1968; Gifford, 1976). It was determined that although the area source emissions constituted only 3.5% of the total area and point source emissions, estimated concentrations from area sources ranged from 14 to 67% of the total concentration estimated at the monitoring sites. For the 11 sites, it was found that CDM slightly over-predicted concentration values by a factor of 1.10 with a correlation coefficient (r^2) equal to 0.96. Nine of the 11 sites have estimates within a factor of 2, with 3 values within 30% of those observed.

The version of CDM applied by Irwin and Brown is similar to the Industrial Source Complex Long-Term (ISCLT) model (U.S. Environmental Protection Agency, 1995). The ISCLT area source algorithm nearly approximates what is obtained when one computes area source impacts using an hour-by-hour simulation (which employs a double integral over the area and hence is currently our best expression of dispersion from an area). The emphasis on improving the treatment of area source impacts reflects the recognition that area source emissions (if present) often

account for a major portion of the simulated impacts. The primary focus of Irwin and Brown was to investigate the sensitivity of the annual concentration estimates to the resolution employed in defining the area source emissions. Within the central part of St. Louis, 0.5 by 0.5 km grids had been used to define the emissions, whereas in the suburbs, the emissions were defined using grids as large as 10 to 20 km on a side. It was found that any reduction in the resolution, say by redefining the emissions into grids of 1 km on a side, significantly reduced the concentration estimates, and caused CDM to underestimate the annual average concentration values (especially in the central portion of the city).

In the studies summarized, it is important to remember that the long-term models have evolved from first generation to second generation models, with the adoption of improved characterizations for plume rise, plume dispersion, and treatment of area sources. Except for the simulations for Nashville by Pooler and for St. Louis by Martin and Calder, the average bias has been slight, with typically 80 to 90% of the estimates being within a factor of two of those observed. This model performance was achieved with research grade emission inventories that had little to no bias. The lesson to be learned from modeling studies of annual or seasonal concentration values is that the skill in the modeling results is typically at the mercy of the diligence employed in specifying the diffuse low-level emissions, as they can dominate the analysis.

The results by Gifford and Hanna (1973), Hanna et al. (1982) and Irwin and Brown (1985) confirmed the importance of specifying these diffuse low-level emissions (typically characterized using area sources) with as much care and resolution as feasible. Interestingly, this lesson is rarely considered when emission inventories are developed. The tradition in inventory development is to estimate the total mass of emissions. This means that the low-level diffuse emissions, which may represent less than 10% of the total mass of emissions, receive the least attention and quality control, even though they may account for as much as 60% of the observed impacts on nearby receptors. This becomes of particular concern when attempting to characterize impacts from species directly emitted into the atmosphere. The uncertainty in distributing (in time and space) the low-level emissions ultimately defines the lowest resolution possible in the modeling assessment.

The long-term urban-scale models provided the first demonstrations of how air quality modeling could be used in the development of emission control strategies for state implementation plans. They also provided an important lesson on properly estimating the spatial distribution and temporal behavior of low-level diffuse emissions, especially for estimation of human exposures from these low-level emissions.

5 Development of Tropospheric Chemistry Models

From a review of the historical record, we see that sulfur was recognized or suspected as the basis of many disastrous episodes that focused attention on air pollution as a health problem: December 1930 in Meuse Valley, Belgium; October 1948 in Donora, Pennsylvania; 1952 and 1956 in London, England; and November 1953 in New York, New York (Stern, 1977). The local and urban-scale plume dispersion models discussed in sections 3 and 4, either ignored chemical transformations or treated their effects as a first-order linear decay. These models were primarily developed for use in control programs to reduce sulfur emissions. By the 1960s, ozone was becoming to be recognized as the basis of the smog events first made famous in Los Angeles, which are now seen not only in California but also from Texas through Georgia, and in all the states along the eastern shore of the United States. In the late 1960s, Europe began a concerted attack on resolving the transport and chemistry of sulfur emissions; whereas, the United States began a concerted attack on resolving the transport and chemistry of ozone. Interestingly, as these investigations matured, each became aware that regional transport and fate could not be ignored, and in many instances was the dominant scale to be modeled. By the mid-1970s, concerns began to be raised that the EPA's program to control Total Suspended Particulate was not sufficiently reducing health risks from particulates. The true problem was perceived to be the very fine particles that could be easily trapped within the lungs.

The development of air pollution models that account for atmospheric chemistry and the formation of secondary products adds several complicating factors to the modeling process, which must be addressed in some manner, or the model simulations are stymied. One must have a model for the chemical kinetics. The reaction rates are determined not only by the availability of the proper chemical constituents, but are also typically functions of air temperature, humidity, and incoming solar radiation. As a further complication, the chemistry is rarely of the elementary variety, but involves a system of coupled reactions that in theory might involve hundreds of relationships, but in practice must be simplified and parameterized to a smaller resolved set based on computational resources. Air pollution chemistry often involves characterizing the time and space variations of the emissions of several chemical species over a broad area from many sources, whose individual contributions are incrementally small compared to the total (from all sources). This is a massive job fraught with uncertainties, that even today have yet to be well addressed. As the computation domain increases in size, simple terrain and land use characterizations must be replaced with more comprehensive characterization of the variations in terrain elevation and land cover, and the effects such variations induce on the local meteorology and emissions. The combination of these factors results in simulation models that are computationally more demanding, which adds a further complication of attempting to simplify the problem without losing some sought after quality. Finally, the nonlinear pathways for the formation and destruction of chemical species make the assessment of model performance through a direct comparison of final products, say ozone

observed versus ozone predicted, problematic. The model has many ways to provide a seemingly correct answer for all the wrong reasons and, if this were happening, it would negate the usefulness of the model to determine control strategies.

The following historical review of EPA's development of tropospheric air pollution models is divided along regulatory and legislative programs. Section 5.1 provides a review of the development of models for EPA for characterization of tropospheric ozone[23]. The idea of controlling tropospheric ozone using local controls of precursor emissions is eventually seen to be insufficient. Section 5.2 provides a review of the development of models for the characterization of acid deposition. Here, the lessons learned in Europe and in the ozone model development program immediately focus on regional transport. The next three sections focus on the 10-year period of 1990-2000. Section 5.3 reviews the development of a model to characterize the transport and fate of fine-particulates (aerosols). Section 5.4 reviews the development of models for characterization of toxic impacts to Great Waters as required by the CAAA of 1990. Section 5.5 reviews the renovation of computer modeling, stimulated by the High Performance Computing Act of 1991, which assisted the EPA in laying the basis for development of a one atmosphere air quality model.

5.1 Ozone

Los Angeles suffered smog events as early as 1903, when the industrial smoke and fumes were so thick that residents mistakenly believed an eclipse of the sun was happening. The smog on July 26, 1943 is often reported as the first recorded episode. After 1943, the frequency of these smog events increased, but the causes were unknown. On June 10, 1947, California signed into law an Air Pollution Control Act[24], authorizing the creation of county-level Air Pollution Control Districts (Los Angeles County was first to create a county-level Air Pollution Control District). These events have bearing on the development of tropospheric chemistry models of air pollution, because it was field data collected in Los Angeles over a series of years that provided a basis for Haagen-Smit (1950) to be able to show that photochemical reactions were the source of the Los Angeles smog. Then Haagen-Smit (1952) was able to show that a mixture of nitrogen dioxide and certain hydrocarbons in air yields ozone in the presence of sunlight. The chemistry kinetics of the reactions were not yet well known, but experiments by Haagen-Smit and Fox (1955) seemed to show that for Los Angeles, the number of ozone molecules seemed to be proportional to the product of the number of

---

[23] See http://www.ksg.harvard.edu/gea/pubs.htm for a review of the legislative and model development for assessment of tropospheric ozone in both the US and Europe by T.J. Keating and A. Farrell (1998). "Problem Framing and Model Formulation: The Regionality of Tropospheric Ozone in the United States and Europe". Belfer Center for Science and International Affairs (BCSIA) Discussion Paper E-98-11, Cambridge, MA: Environment and Natural Resources Program, Kennedy School of Government, Harvard University.

[24] See http://www.arb.ca.gov/html/brochure/history.htm.

molecules of nitrogen dioxide and hydrocarbons. We know now that this characterization is a gross simplification, but interestingly enough, Frenkeil (1957) used this model of ozone production to show that the contribution of each pollution source to the ozone concentration is not directly additive. The sum of the individual contributions was less than the ozone produced when all sources were simulated simultaneously. It appears that F.N. Frenkiel's simulations may be the first air pollution computer simulation, and his results may be the first to comprehensively demonstrate the complexity that chemistry introduces into assessing the effect of different control strategies.

For the 20-year period from Haagen-Smits' early papers on ozone formation to the mid-1970s, the conventional wisdom was that tropospheric ozone formation could only take place in atmospheric environments that were heavily polluted with automobile exhausts and strongly illuminated with sunlight. Leighton (1961) provided a comprehensive analysis of the known principal reactions in smog formation during this period. The next advance was to realize the importance of the role of hydroxyl radical (Levy, 1971, 1972), and the interplay with methane and carbon monoxide (Crutzen, 1973, 1974). The chain of reactions and list of products were expanding rapidly. Several attempts were made by California consulting firms and universities to develop partial photochemical air quality models during the early 1970s, for example: a three-dimensional Eulerian model was developed by Roth et al. (1971) and Reynolds et al. (1973) of Systems Applications, Inc.; a single-moving cell model by Weisburd et al. (1971) and Wayne et al. (1973) of Systems Development Corporation; a Lagrangian column-of-cells model by Eschenroeder and Martinez (1971) and Eschenroeder et al. (1972) of General Research Corporation; and a particle-in-cell model by Sklarew et al. (1971) of Systems, Science and Software, Inc. Each of these were attempts to grapple with the dilemma of sacrificing some aspect of the problem, in order to have reasonable run times and confine the numerical computations to be within the memory limits of the available computers.

EPA was funding the early photochemical model investigations, but the resulting models found only limited use by regulatory agencies. A statistical relationship had been derived - between the 6:00 to 9:00 AM ambient total and non-methane hydrocarbon concentrations, and the corresponding daily maximum ambient ozone concentration - using data from the Continuous Air Monitoring Project (CAMP stations) and Los Angeles during the late 1960s (Schuck et al., 1970; U.S. Environmental Protection Agency, 1971). This was summarized in the 1971 Appendix J to Title 40, Part 51 of the *Code of Federal Regulations*, and was used to estimate the degree of reduction in hydrocarbon emissions (total and non-methane) needed to achieve the primary NAAQS for photochemical ozone (1-hour average of 0.08 ppm, not to be exceeded more than once per year at any one location). One of many shortcomings of the approach was its inability to address transport effects, such as entrainment of unscavenged ozone, which can result from transport from other regions or from ozone trapped aloft overnight.

The first reports on photochemical smog in London and Western Europe were presented by Derwent and Stewart (1973) and Guicherit and van Dop (1977). By 1977, EPA had completed its efforts to find a replacement for the 1971 Appendix J procedure. Based on smog chamber studies, a chemical kinetics model was derived for the formation of ozone from a mixture of propylene, n-butane and nitrogen oxides. This kinetics model formed the basis of the EPA Empirical Kinetic Modeling Approach (EKMA), which utilized a set of empirical ozone isopleths depicting the maximum afternoon ozone concentration downwind of a city as a function of initial morning concentrations of precursor emissions (non-methane hydrocarbons and nitrogen oxides), precursor emissions occurring later in the day, meteorological conditions, reactivity of the precursor mix, and concentrations of ozone and precursors transported from upwind areas. EKMA was a single-column box model that could be envisioned as following a trajectory. As the column of air is advected with the winds, emissions that enter are assumed to be instantly mixed uniformly within the column. From early morning to mid-afternoon, the column height increases to simulate the growth of the mixing height during the day and effects of entrainment of pollutants trapped aloft (Dodge, 1977). This model contained a detailed characterization of ozone formation and fate, and retained computational efficiency. For the next 10 years, EKMA found widespread use to predict the relative changes needed in the precursor emissions necessary to reduce observed maximum ozone concentration for an area to be below the new 1-hour maximum ozone NAAQS of 0.12 ppm.

EPA had recommended two approaches for formulating State Implementation Plans to achieve the NAAQS for ozone. The first was EKMA that allowed development of city-specific plans. The second approach allowed the use of a grid-based photochemical air quality model, which by necessity used a reduced (or lumped) chemical model. From 1977 to 1987, the EPA sponsored research resulted in two reduced mechanisms: the lumped molecule (surrogate species) approach (Carter et al., 1986; Lurmann et al., 1987) and the lumped structure (Carbon Bond I through IV) approach (Whitten et al., 1980; Gery et al., 1988). The critical review by Seinfeld (1988) described the evolution of the first-generation grid models towards becoming second-generation models, and also summarized several of the practical implementation problems of the grid models, which primarily resided in the considerable database needed for their use. He also noted that rigorous, universally-accepted performance criteria for grid-based photochemical air quality models do not exist, a statement that is still true in 2005.

For regulatory applications, EPA released version 2 of the Urban Airshed Model (UAM-II) in 1980 (U.S. EPA, 1980) and later recommended it as the preferred model for ozone planning in urban environments (U.S. EPA, 1986). In 1988, improvements were made to the chemical mechanism and a system of preprocessors for preparing emissions and meteorological inputs were added to create a second-generation modeling system, UAM-IV (Gery et al., 1988). As the limitations of EKMA became recognized, UAM-IV began to emerge as the dominant tool for urban-scale ozone planning.

As described by Strothmann and Schiermeier (1979), a White House initiative in late 1971 called for development and validation of improved air quality simulation models upon which cost-effective pollutant control strategies could be based. This provided the basis for EPA's St. Louis Regional Air Pollution Study (RAPS). Planning for this study began in August 1971, and field investigations were conducted approximately between 1973 and 1978. One of the primary objectives of RAPS was to create a comprehensive and accurate database for all criteria pollutants and selected non-criteria pollutants, for use in developing and evaluating air quality simulation models, with particular emphasis on photochemical models. For this purpose, the Regional Air Monitoring System (RAMS) was designed and operated, which consisted of 25 remotely operated, automated monitoring stations controlled and polled via telemetry by a central data acquisition system. Station locations were chosen with care to avoid being unduly affected by emissions from some local nearby source. Comprehensive point and area source emission inventories were developed including emissions of sulfur oxides, carbon monoxide, oxides of nitrogen, hydrocarbons and particulates. Seventeen stations had 30-m meteorological masts while the other sites had 10-m masts. The meteorological instrumentation was comprehensive, including wind sensors, temperature, dew point, pressure, and solar radiation. In addition, the 30-m masts had a 5 to 30 m temperature difference measurement, and a UVW Gill anemometer for turbulence measurements. An upper air-sounding network was established to provide a definition of the winds and temperature structure aloft. A review of the many publications spawned by this study reveals that they fall into two basic categories: studies of urban air quality and meteorology (e.g., Ching and Doll, 1981; Clarke et al., 1981; Godowitch et al., 1979, 1981; Karl, 1980; Shreffler, 1978) and studies of air quality model performance (e.g., Schere and Shreffler, 1983; Turner and Irwin, 1983, 1985; Turner et al., 1985).

EPA-sponsored field studies (e.g., U.S. Environmental Protection Agency, 1976) revealed the regional nature of the ozone problem. It was becoming increasingly clear that local reductions in ozone precursor emissions were an insufficient abatement strategy for many areas of the U.S. Crafting an effective ozone reduction program would require following the fate of air masses for several days, or in other words, a regional abatement strategy. In 1977, EPA began the development of the Regional Oxidant Model (ROM) (Lamb, 1983, 1984) which found extensive use in the examination of the effects of alternative emission reductions on ozone concentration for the eastern United States. The chemistry kinetic mechanism used in ROM was the lumped structure approach (Carbon Bond I through IV). To conserve computer memory, the atmosphere was vertically divided into three layers: layer 1, the surface-layer and lower boundary layer; layer 2, a layer whose top followed the diurnal extent of the mixed layer; and layer 3, an upper layer capping inversion layer. The wind fields were determined by interpolation on an hour-by-hour basis from observations at surface and upper air stations. The strength of this model was its ability to follow the chemistry of 28 species, and cover almost one-fourth of the United States in one run. The

limitations were the complexity of model setup, and use of boundary conditions[25] tuned to particular locales through extensive testing, which precluded recommending unassisted use of the model for compliance modeling (Schere and Wayland, 1989).

In 1991 the U.S. National Research Council, with funding support from the EPA, completed its scientific assessment of the state of knowledge of the physical and chemical sciences relevant to the characterization of tropospheric ozone (National Research Council, 1991a). They concluded that despite a major regulatory effort, the ozone control programs for the 1970 to 1990 period had failed. The State Implementation Plans were fundamentally sound as planning instruments, but seriously flawed due to the lack of a verification program. Emission inventories were seen to be highly uncertain, with anthropogenic volatile organics significantly underestimated, and they saw a need to recognize the important role of biogenic volatile organic emissions in simulating tropospheric ozone. The National Research Council review also noted that despite over a decade of experience in ozone modeling, there had yet to be a consistent set of evaluation procedures, notwithstanding the widely recognized need for such methodology. At this point in time, the UAM model with Carbon Bond II was in wide use. There had been some testing in several models with Carbon Bond IV kinetics (CBM-IV). The software package that allowed EKMA control strategies to be estimated using optional chemical kinetics models was upgraded for a fourth time to include CBM-IV and a feature that allowed a user-defined mechanism, OZIPM-4 (U.S. Environmental Protection Agency, 1988a). Recognizing the importance of regional transport, U.S. Environmental Protection Agency (1988b) issued guidance that suggested that UAM might be used to provide the regional boundary conditions for use in the OZIPM analyses.

In 1991, the results and conclusion of a 3-year study called the Regional Ozone Modeling for Northeast Transport (ROMNET) program were published (Possiel et al., 1991). This investigation used ROM version 2.1, employing meteorological scenarios from episodic periods in 1980, 1983, 1985 and 1988, with emissions for each episode and five control strategy projected emission inventories for each episode for 1995 and 2005. A major conclusion was that the areas outside of the corridor of cities along the east coast of the United States may have to add controls beyond those necessary to solve their local problems in order to reduce transport of air pollutants and precursors into these areas. For most of the domain, the production of ozone was seen to be limited by the oxides of nitrogen, with ozone production limited by the availability of volatile organic compounds in areas with large oxides of nitrogen emissions. Meteorology played a major role in the buildup and limitation of regional ozone. These results confirmed the important regional nature of the ozone formation and the need for development of regional air

[25] Generally, ROM was run with tropospheric clean boundary conditions, and EPA's Aerometric Information Retrieval System (AIRS) ozone measurements could be used for continental side boundaries (http://www.epa.gov/airs/airsaqs/).

pollution control programs. These ROMNET results were confirmed in a later investigation that employed ROM version 2.2 (Roselle and Schere, 1995). In this later investigation, a 9-day period in July 1988 was simulated for the eastern US, with 17 simulations of various control strategies. The improvements made in going from ROM version 2.1 to version 2.2 included a well-mixed convective boundary layer model, improved characterization of wind flow during nighttime inversion conditions, upgraded characterization of turbulence over urban areas, and upgraded parameterizations of the vertical cumulus cloud flux. There were other studies as well, and it was seen that advances in scientific understanding were not altering our growing awareness of both the regional nature of ozone formation and the general conclusions found in ROMNET. All of the studies highlighted need an operational regional ozone model for routine use. In 1996, the decision was made that all subsequent oxidant model development would be conducted within the one atmosphere paradigm, which at the time was called Models-3/Community Multi-Scale Air Quality (CMAQ) modeling system.

There was a sense, left by the 1991 National Research Council review, that the ozone models in routine use were need of revamping. The modeling tools under development were admittedly pushing the limits of the computers of this time. Expanding the scope to include regional transport would only exacerbate the problems imposed by the computer limitations. The ozone standard and the effectiveness of the control programs were in question. We close this section of the discussion with the recognition that the 1990 decade is the advent of third generation models, which will require redesign of the numerical framework, the chemistry mechanism, fuller characterization of mesoscale meteorological effects, and consideration of the possibility of feedbacks between the chemistry, meteorology, and surface heat and moisture balance.

5.2 Acid Deposition

Even though our emphasis is regulatory model development for EPA, it is worthwhile to examine Europe's interest in air quality modeling of sulfates for two reasons. First, a comparison of Europe's early models for sulfate reveals that they are conceptually similar to the designs used in the US for early ozone models. Second, when the US finally began to focus on modeling sulfates, they borrowed heavily from the experiences of Europe. The period from the mid-1950s through the mid-1970s can be viewed as the development of a conceptual model for the acidification of precipitation in Europe. The acidification of precipitation in Europe was first noticed in samples collected from the European Air Chemistry Network, established in the mid-1950s (Ottar, 1978). Analyses of these data showed that in Europe, the central area with highly acid precipitation was expanding. The main acid component was sulfuric acid, and was seen to be related to the increasing use of sulfur-containing fossil fuels in Europe. The Organization for Economic Co-operation and Development (OECD) recommended a full investigation. With the approval in 1972 by the OECD, the first measurement program was launched. A second measurement program was completed in 1975.

By 1977 it was becoming clear that even though the countries with the largest emissions also received the largest depositions of acidic rain, those regions with low emissions received more pollution from other countries than from their own sources.

Development of mathematical simulation models in Europe had a decided focus on long range transport effects during the period from 1970 to 1980. The early Lagrangian models followed moving air parcels along trajectories (Eliassen, 1978), and the transformation and deposition were approximated using an assumed rate of transformation of sulfur dioxide to sulfate, and assumed wet and dry deposition velocities for sulfur dioxide and sulfate. The early Eulerian grid models had similar characterizations for the transformation and deposition, and suffered (as all early grid models) from undesired computational dispersion (Nordo et al., 1974). These models could achieve high correlations with annual average concentration values, but were incapable of correlating well with 24-hour averages (or less). Statistical models were developed that were able to describe the broad features of the annual average patterns (Rodhe, 1972; Fisher, 1983). The purpose of these statistical modeling efforts was to confirm the conceptual model that had been constructed from analysis of precipitation records that the developed industrial regions of central Europe were responsible for most of the acidification of the precipitation seen in Norway and Sweden.

A change occurred in the direction of model development and air pollution policy in the United States around 1976. Control of sulfur dioxide had decreased urban levels of sulfur dioxide, but these decreases did not seem to be accompanied by a proportional decrease in urban sulfate, which was popularly called acid rain. Of the existing explanations for the lack of decrease in sulfate levels, the results from European investigations seemed to provide the most plausible theory, that is, the long range transport of sulfur emissions was responsible for the observed urban sulfate levels.

There were significant scientific uncertainties in the health effects and in the available characterizations for long range transformation-transport of sulfate. To provide a scientifically sound assessment of the extent of the acid rain problem, Congress passed and President Carter signed the Acid Precipitation Act of 1980 (Public Law 96-294). The Act directed the United States Government to conduct a ten-year assessment to determine the causes and consequences of acid precipitation, and to develop options for reducing known effects. The National Acid Precipitation Assessment Program (NAPAP) was created in response to this congressional charge. NAPAP formally involved twelve Federal agencies, and informally several states and provinces of Canada, EPRI, the National Association of the Paper Industry for Air and Stream Improvement, the Mellon-Foundation, and the Natural Resources Defense Council. NAPAP was reauthorized for an indefinite period through Title IX of the CAA Amendments of 1990, Cowling (1992). Thus, in the late 1970s and throughout the 1980s, funding for model development for abatement of sulfur dioxide and urban ozone was reduced to

support an aggressive program for development of regional-scale acid precipitation air quality models.

The first of a series of plume mapping and sampling field studies was the Midwest Interstate Sulfur Transformation and Transport (MISTT) from 1973 to 1976 (Wilson, 1978). The electric power utilities sponsored the Sulfate Regional Experiment in Northeastern United States (SURE), which began in 1976 with a network of 54 hi-vol, and sequential samplers stations distributed somewhat randomly throughout the northeastern United States to provide measurements of particle mass, sulfate, sulfite, nitrate, chlorate, ammonium and water-soluble organics (Perhac, 1978). EPA initiated the Sulfur Transport and Transformation in the Environment (STATE) field studies, of which the first was the 1978 Tennessee Plume Study (TPS) (Schiermeier et al., 1979). Using aircraft sampling, the TPS attempted to quantify changes in plume characteristics and composition out to distances of 500 km. The NOAA Air Resources Laboratory coordinated the 1983 Cross-Appalachian Tracer Experiment (CAPTEX), which involved over eighty sequential ground-level tracer samplers of perfluorocarbon at distances of 300 to 1100 km from release sites (Ferber et al., 1986). The upper-air soundings were increased to four per day, and seven aircraft provided vertical distributions of the tracer. This rich database has found frequent use to test a variety of advancements in regional-scale modeling techniques, including: Four Dimensional Data Assimilation (FDDA) (Kao and Yamada, 1988); particle dispersion modeling (Lee, 1987); and delayed shear enhancement (Moran and Pielke, 1996).

In 1982, EPA asked the National Center for Atmospheric Research (NCAR) to assess the state of the sciences that would be involved in developing comprehensive acid deposition modeling systems. This assessment (National Center for Atmospheric Research, 1983a, 1983b) and other independent assessments (MOI, Work Group II, 1982; Electric Power Research Institute, 1984) concluded that it was feasible to develop a comprehensive wet and dry acid deposition model. In 1983, the Acid Deposition Modeling Project was established at NCAR to develop a model suitable for use by NAPAP. The project was funded by the EPA and the National Science Foundation (NSF), and it was based at NCAR until 1987. With the shift in 1987 towards model testing and application, the project was moved to the Atmospheric Sciences Research Center (ASRC) of the State University of New York (SUNY) in Albany, NY. Several versions and enhancements were made to the Regional Acid Deposition Model (RADM) as a consequence of this work. RADM was a numerical grid model that subdivided the northeastern United States into a 35 by 38 horizontal grid with six to fifteen vertical levels. Aqueous-phase reactions in clouds are a major contributor to atmospheric acidification; therefore, it was important to employ a chemistry kinetic mechanism that could correctly predict the concentration, solubility, and rate of mass transfer of oxidizing agents such as hydrogen peroxide, ozone, methyl hydrogen peroxide, peroxy acetic acid, and hydroxyl and hydroperoxy radicals. With these concerns in mind, the RADM organic chemistry mechanism was developed using a reactivity lumped molecule approach. By 1990, the gas-phase chemical reaction mechanism

contained over 100 reactions and followed over 50 species. New methods were found to limit numerical solver uncertainties; enhancements were made to better address aqueous-phase in-cloud chemistry and dry deposition fluxes were computed for 13 species (Chang et al. [1990], 4-113).

The abundance of naturally occurring hydrocarbons in the atmosphere had been recognized for some time (Arnts and Meeks, 1981; Peterson and Tinge, 1980; Rasmussen, 1972). Even before modeling studies by Chameides et al., 1988 and Trainer et al., 1987 were suggesting the need to consider biogenic hydrocarbon emissions for estimating the production of photochemical oxidant, EPA reported on the development of a computer algorithm for estimating biogenic emissions (Novak and Reagen, 1986). This system, called the Biogenic Emissions Software System (BESS), was designed to produce hourly gridded hydrocarbon emissions for an early version of the ROM. In parallel with this effort, researchers at Washington State University devised a method for estimating seasonal, county-wide hydrocarbon emissions across the US (Lamb et al., 1987). These estimates were used for early parts of the NAPAP. These two methods for estimating biogenic emissions were combined by Young et al. (1989), which became a generalized scheme called the Biogenic Emissions Inventory System (BEIS) that could service either ROM or RADM (Pierce et al., 1990).

The meteorological model for RADM was the Pennsylvania State University/National Center for Atmospheric Research (PSU/NCAR) Mesoscale Model, Version 4 (MM4). EPA initiated work towards development of this model in the early 1970s with a research grant to the Pennsylvania State University (Anthes et al., 1974; Anthes and Warner, 1978; Anthes et al., 1987). This research involved converting a mesoscale meteorological model that simulated hurricane dynamics where atmospheric processes are strongly forced, into an air quality meteorological model, which often must deal with weakly forced or stagnant conditions. This model was the first to employ Newtonian Relaxation, or nudging, in which the model state was relaxed toward the observed state by adding to one or more of the prognostic equations, artificial tendency terms that were based on the difference between the two states. In making the conversion for air quality modeling, the nudging was extended to include surface boundary layer variables (Stauffer and Seaman, 1990; Stauffer et al., 1991).

The 1980 charge to perform a comprehensive scientific assessment of the fate of sulfate and the aggressive response by NAPAP to form a comprehensive regional-scale acid precipitation model accelerated the advancement of chemical solvers, the characterization of meteorological transport and atmospheric processes. The demands for more complete characterizations of the chemistry placed increasing demands for detailed comprehensive emission inventories. For instance, the 1990 version of RADM 2.0 required hourly emissions of sulfur dioxide, sulfate, nitric oxide, nitrogen dioxide, ammonia, carbon monoxide, and 15 classes of volatile organic compounds (VOCs) (Chang et al. [1990], 4-27). The Acid Precipitation Act of 1980 and the resulting creation of the NAPAP caused an

intersection between the critique of what science can affirm and the development of policy. One of the many lessons learned was that science cannot answer policy questions, which are value-based; whereas science can provide an assessment of the consequences of alternative strategies. This was not an easy intersection as witnessed by the comments and reflections of those involved (Cowling, 1992; Kingdon, 1995, Lackey and Blair, 1997; Alm, 2000).

The RADM NAPAP assessment was completed in 1990 with significant participation by many ARL NOAA scientists (Hicks et al., 1990; Binkowski et al., 1990; Chang et al., 1990; Dennis et al., 1990a, b). Of the many advances made in developing and applying RADM, of significance are: 1) a dynamic mesoscale meteorological model (MM4) was successfully tailored for use in air quality simulations by extending FDDA to include nudging of winds and moisture within the planetary boundary layer; 2) MM4 was successfully coupled to a dynamic air quality model (RADM) with a comprehensive characterization of oxidant and sulfate chemistry, and fate; 3) the importance on air quality modeling results of the simulation of cloud fields, cloud processes, and heterogeneous and aqueous chemistry was successfully demonstrated; 4) an aggregation methodology was successfully developed that allowed long-term concentration averages to be estimated using a combination of short-term episodic results (Samson et al., 1990); and 5) a series of engineering versions of RADM were successfully developed that allowed rapid and accurate investigation of sulfur control strategies without the need to make additional runs of the resource and computationally demanding RADM.

In the years following the completion of the RADM evaluation and its use in the NAPAP assessment, there were several important advances that were tested and evaluated. A simple non-local closure model for characterizing vertical mixing within a convective boundary layer (CBL) (Pleim and Chang, 1992) addressed the findings of the convective tank experiments by Willis and Deardorff (1976, 1978, 1981) within the context of an Eulerian grid model. The model, named the Asymmetrical Convective Model (ACM), allows mixing from the lowest model layer directly to all other layers in the CBL, but restricts downward transport to proceed only to the next lower layer, in order to emulate the rapid upward mixing of convective eddies, and the much slower downward mixing typical of convective boundary layers. They installed ACM within RADM and saw that the rapid transport of surface emissions of nitrogen oxide and nitrogen dioxide into layers aloft coupled with the slower downward mixing resulted in lower ozone concentrations throughout the mixed layer than would be obtained using conventional vertical mixing models. In 1993 and 1994 (Poole-Kober and Viebrock, 1993, 1994), ARL staff performed several in-house investigations to better understand how grid resolution in the vertical and horizontal affected RADM model performance. It was concluded that increasing the vertical resolution from 6 to 15, and then to 30 layers improved the simulation of nocturnal surface concentration values of ozone, because the oxides of nitrogen emissions emitted into a thinner lowest layer were less diluted, and this enhanced the titration of

ozone at night. In a 1994 investigation, ARL staff conducted in-house sensitivity analyses using RADM to test the potential of two heterogeneous reactions on oxidant photochemistry. One was a heterogeneous reaction of N_2O_5 with water to produce nitric acid, and hence, the termination of photochemical active oxides of nitrogen. The other was the conversion of hydroperoxy radical to hydrogen peroxide. There was evidence that this reaction occurs on wetted aerosols when aqueous copper concentration is sufficiently high to act as a catalyst. Investigations concluded that the first reaction could be included in an operational RADM, but the second reaction relied on copper content estimates of aerosols, which was considered to be too uncertain for operational use.

In 1993, a one-dimensional prototype program was developed that consisted of a simple surface energy and moisture parameterization including explicit representation of soil moisture (Noillian and Planton, 1989) with the ACM model. The coupled surface and planetary boundary layer (PBL) model performs integrated simulations of soil temperature and soil moisture in two layers as well as PBL evolution, and vertical transport of heat, moisture, and momentum within the PBL. Comparisons of modeling results with observations taken during a two-day period of the Wangara field study and several days from the First ISLSCP Field Experiment (FIFE) in 1987 and FIFE in 1989 illustrated the model's ability to simulate ground temperature, surface fluxes, and boundary layer development accurately (Pleim and Xiu, 1993). This model was then incorporated into MM4, essentially replacing the existing high resolution PBL model, and work was initiated to develop an advanced FDDA technique for indirect nudging of soil moisture. By 1996, the model had been installed within the newly released MM5 (Grell et al., 1994) and was denoted as MM5PX. There were several improvements provided in MM5, the main one being that it contained both the hydrostatic and non-hydrostatic equations of motion. Indirect nudging of soil moisture was seen to explain errors in simulated air temperature, humidity and PBL heights (Pleim and Xiu, 1995). Collaborative research with NCAR converted this surface model for inclusion in MM5 version 3, which was released in July 2000.

It is 1996 at this point in the discussion, and the decision is made that all subsequent acid deposition model development would be conducted within the one atmosphere paradigm, which at the time was called Models-3.

5.3 Aerosols

In 1987, early models of the three general approaches available for the characterization of the aerosol distribution were being investigated (Hudischewskyj et al., 1987). The three approaches involved a continuous representation (Suck and Brock, 1979; Tsang and Brock, 1982, 1983), a sectional representation (Gelbard and Seinfeld, 1980; Seigneur, 1982, Gelbard, 1984; Warren and Seinfeld, 1985), and a modal representation (Whitby, 1978, 1981, 1985; Saxena et al., 1986). The modal model represents the entire fine aerosol distribution as the summation of two log-normal distributions. The processes that

affect the aerosol distribution are simulated by altering the zero-th, third and sixth moments of these two distributions. The modal representation was determined to be on average 400 times faster with average errors for nitrate, ammonium, total mass, and the light scattering coefficient (b_{scat}) all well within 10 percent in comparison with the other two representations. With the completion of the Modal Aerosol Dynamics (MAD) model, (Whitby, 1990; Whitby et al., 1991), developmental work was initiated to install the MAD model into RADM. A technique for estimating dry deposition fluxes for particulate matter was developed that accounted for the particle size-dependent effects of Brownian motion, inertial impaction, and gravitational settling (Bullock, 1990). Comparisons with measured field data (Wesely et al., 1985) provided a basis for selecting a deposition velocity characterization by Pleim et al., 1984. By 1994 the focus was on improving the characterization of cloud interactions on aerosol size dependence and on the characterization of wet deposition (Binkowski and Shankar, 1994; Shankar and Binkowski, 1994). In 1995, the number of chemical aerosol species was increased by adding nitrate and organic carbon to the existing list of sulfate, ammonium, and water.

The first generation Regional Particulate Model (RPM) was formed in 1989 by adding an aerosol chemical and kinetic mechanism to RADM/EM to characterize the development and fate of aerosols. This model was then capable of simulating sulfate aerosols in seven size ranges (a sectional approach), and considered the effects of nucleation, condensation, evaporation, coagulation, and aqueous aerosol chemistry. This model was developed to assist in policy development and promulgation of air quality standards for fine particles, visibility, and acid aerosols (Bullock et al., 1989). Preliminary review of predicted sulfate concentration values for August 3-6, 1979 of the Northeast Regional Oxidant Study (NEROS), and for April 22-24, 1981 of the Oxidation and Scavenging Characteristics of April Rains (OSCAR) tended to be highest where observed visibility was lowest. Aerosol concentration values near the source regions tended to be low, with higher aerosol concentration values further downwind of the source regions.

In 1995, a new method for modeling dry deposition of gaseous chemical species was developed to take advantage of the more sophisticated surface model implemented in MM5PX (Pleim et al., 1996, 1997), and was tested against field data observations for ozone deposition. Since MM5PX had a parameterization for evapotranspiration, the same stomatal and canopy conductances could be used to compute dry deposition velocities of gaseous species. While developing the model, it was seen that it could be easily extended for use to compute dry deposition velocities directly from field measurements (Pleim et al., 1999). The scheme potentially provides a means for accurately estimating the dry deposition velocity of ozone, and perhaps other gaseous species such as sulfur dioxide and carbon monoxide, from relatively inexpensive field networks and without the need for direct chemical eddy correlation measurements.

By 1996, the RPM had become a complete aerosol and visibility modeling system (Binkowski and Shankar, 1995; Binkowski and Ching, 1996), and the decision was made that RPM would be incorporated into the Models-3/CMAQ framework.

5.4 Toxics

Title III of the CAAA of 1990 required an assessment of the annual atmospheric deposition of toxic substances to the Great Waters that consist of the Great Lakes, the Chesapeake Bay, Lake Champlain, and the coastal waters of the US. This included the identification of the sources and assessment of their relative contributions. To provide an interim assessment using off-the-shelf technology, it was decided to adapt an existing model, called RELMAP (Eder et al., 1986), to simulate atmospheric deposition of toxic substances (Clark et al., 1992; Clark, 1992). It was assumed that the substances are chemically inert and deposit at rates based on published physical attributes (i.e., Henry's law coefficients, liquid-phase vapor pressures, and phase partitioning ratios). Using existing emission inventories, it was possible to simulate 22 toxic substances, and the preliminary results confirmed that the transport scales of the substances deposited to Lake Michigan varied significantly. In 1993, this effort focused 1) on the characterization of mercury emissions from all major anthropogenic sources, and 2) on mercury emissions and other designated toxic pollutant (arsenic, cadmium, lead, and 17 separate dioxin/furan congeners) only from coal-fired utilities. The latter effort culminated in a final mercury study report to the U.S. Congress (U.S. Environmental Protection Agency, 1997) and in a final report to the U.S. Congress on the hazardous air pollutant study for the electric utility steam generating units (U.S. Environmental Protection Agency, 1998a). Recent laboratory studies of chemical reactions of mercury and its compounds in air and in water (Lin and Pehkonen, 1999) suggest that the chemistry mechanism of RELMAP mercury may not accurately reflect the complex nature of mercury chemistry, especially in cloud water. In response to this, work has begun to modify CMAQ (discussed in section 5.5) to include mercury and various mercury mechanisms as modeled species. One of the serious uncertainties to be addressed in this work will be estimating the concentration of chloride ions in cloud water from marine to continental locations.

It was realized, at the inception of the work effort just described, that there were limitations inherent in the RELMAP modeling system for the treatment of chemistry and cloud processes. Hence in 1991, the EPA Office of Research and Development and the Air, Radiation and Toxics Division of EPA Region III created the Chesapeake Bay Evaluation and Deposition (CBED) Committee. This second effort was to have a research focus, but also serve a practical regulatory goal of investigating what reductions might be expected in nitrate deposition to the Chesapeake watershed and to the tidal Bay, resulting from control programs brought about by the CAAA of 1990 to reduce ozone. In particular, the Chesapeake Bay Program jurisdictions had a goal of reducing nitrogen and phosphorus pollution by 40 percent (from 1985 levels) by the year 2000. In the course of this work, it was decided to provide the RADM deposition results to the

Chesapeake Bay Watershed Model (Donigian et al., 1991; Linker et al., 1993). This provided a means for estimating the nitrogen loadings to be expected in the tidal waters of the Bay, based on estimated reductions in atmospheric nitrogen loadings to the Bay waters. By 1996, the focus was on defining scenarios to be simulated by RADM, which would be run with a 20 km grid resolution covering the Chesapeake Bay watershed, and to link these results to the watershed model. These analyses were directed towards assessing the reductions that might be anticipated in nitrogen deposition, resulting from the implementation of three scenarios: 1) the expected 1990 CAAA oxidant-related emission reductions when mandatory control programs are applied; 2) scenario 1, plus a low emission vehicle program and emission limits applied to large fuel combustors in the Northeast Ozone Transport Region, and 3) scenario 2 applied to all states in the Chesapeake Bay watershed. From these results, it appears that scenarios 1 and 2 would reduce nitrogen loadings by about 10 to 15 percent, whereas scenario 3 would reduce nitrogen loadings perhaps as much as 30 percent (U.S. Environmental Protection Agency, 1996; Dennis, 1997). A significant finding in these investigations was the importance of considering the transport of deposited nitrogen by feeder streams that drain into the tidal Bay, which might account for as much as 20 percent of the total nitrogen loading to the tidal Bay, and thus, the benefits of considering control programs that affect the entire watershed system. In 2000, plans were made to use an extended RADM that incorporates the full dynamics of secondary inorganic fine particle formation in order to study ammonia deposition, and to produce revised estimates of the deposition of oxidized nitrogen to the Chesapeake Bay and its environs.

The Chesapeake Bay study provided ample demonstration of the benefits of linking environmental models (air, soil, and aquatic) in assessing pollution impacts. During these investigations it became increasingly apparent that there were many scientific issues and uncertainties in linking together process-oriented models from different media (air, soil, water) that simulate the transport and fate of pollutants. Thus, a long-term research and development project was started in 2000, called the Multimedia Integrated Modeling System (MIMS), whose goal was the development of a modeling system with predictive capability for transport and fate of nutrients and chemical stressors over multiple scales, to allow assessment of air quality and watershed management practices on stream and estuary conditions. The system would involve characterization of chemicals through the hydrologic cycle, or the response of ecological systems to land-use change. Currently, MIMS is not anticipated to couple Models-3/CMAQ within MIMS, but use results from Models-3/CMAQ as input to the MIMS simulations.

There is a class of local-scale modeling that up until this point has not been addressed in this review, and it has to do with what is traditionally referred to as human exposure modeling. Human exposure modeling assessments have had a role in the regulatory assessments leading up to establishing or revising the NAAQS. The goal of such modeling assessments is to characterize the distribution of exposures that the population will experience due to ambient levels of pollutant

concentration. Studies of human time-activity patterns show that people move from various indoor environments to outdoor environments, and the movement involves commuting from one location to another. The term microenvironments is used to denote different places (car, home, office, construction site, restaurant, sports arena, parks, etc.) one might describe as their location during a daily activity record. The time one spends in each of these microenvironments is a function of age and occupation. Modeling exposures of populations (age-sex-occupation subgroups or cohorts) then becomes combining the microenvironmental concentration with time-activity patterns specific to each cohort, and extrapolating these results to include the entire population (Duan, 1981, 1989). An early successful model of this form for simulating personal exposures, called SHAPE (Ott et al., 1988), was evaluated for carbon monoxide exposures using personal exposure data collected in Denver, CO (Johnson, 1983). These investigators have since extended these modeling concepts to allow the investigation of how much of PM10 personal exposures can be attributed to ambient emissions versus indoor emissions (Ott et al., 2000).

Data on human activity patterns were combined with measurements of ambient concentrations using the NAAQS exposure model (NEM) (Biller et al., 1981) to estimate population exposures, and to assess the effectiveness of proposed rulemaking involving particulate matter (Johnson and Paul, 1981), carbon monoxide (Johnson and Paul, 1983) and nitrogen dioxide (Johnson and Paul, 1984). To improve the characterization of variability possible in the population exposures, the NEM modeling assumptions regarding air exchange rates between microenvironments, generation of time-activity patterns, and certain other empirical parameterizations were converted from being average characterizations to probability distributions. This second-generation NEM (pNEM) was then used to assess carbon monoxide exposures to Denver residents (Johnson, 1992) and ozone exposures in the Chicago urban area (McCurdy, 1994). In accordance with a peer review recommendation that pNEM be evaluated against actual personal exposure data, an evaluation was conducted in which pNEM estimates of carbon monoxide were compared with carbon monoxide observations (Law et al., 1997) available from a personal exposure monitoring study conducted in Denver (Johnson, 1983). The evaluation results suggested that pNEM was underestimating the highest personal exposures and overestimating the lowest personal exposures.

Using concepts from the probabilistic NEM model, a Hazardous Air Pollutant Exposure Model (HAPEM) was streamlined and specialized for use in simulating exposures from mobile source emissions (Johnson et al., 1992), and the model came to be called HAPEM-MS. Of significance was the fact that the model and its required databases were revised so that the model could be run on a desktop computer rather than a mainframe computer. HAPEM-MS was then used to estimate carbon monoxide exposures for Denver (Glen and Zelenka, 1994). In 1998, HAPEM-MS was used to produce the exposure estimates for the National Air Toxics Assessment for the base year of 1996. This assessment produced results that are useful in understanding the quality of air and its possible effect on human

health nationwide. Specifically, it provided estimates of 1) the release of 33 toxic atmospheric compounds into the air from various sources; 2) the concentration of these compounds in air; 3) the exposure of populations to this air; and 4) the risk of both cancer and non-cancer health effects resulting from this exposure. Work has continued on this through 2000, and it is expected that additional years will eventually be simulated[26].

The preceding discussion of pNEM and HAPEM-MS described population exposure models that assume that either monitoring data or model estimates are available for characterizing the ambient concentration values. In 2000, a new project was initiated with a goal of developing algorithms for linking Eulerian grid-based air quality models to human exposure models to provide a means for understanding, quantifying and assessing the health impacts and risks of airborne particles and air toxics. In designing these algorithms, it is anticipated that details of the concentration variation will require algorithms that directly or indirectly estimate the subgrid neighborhood-scale variation in the concentration values. To initiate this project, an Eulerian grid model (CMAQ, discussed in section 5.5) was run for the Philadelphia Metropolitan Area for three grid resolutions (12, 4 and 1.33 km) to investigate the sensitivity of the simulated surface layer ozone and PM2.5 concentrations to grid resolution. Future investigations are needed to develop methods not only for assessing the practical limits of grid modeling, but also for addressing urban canopy effects, exchanges between microenvironments, and methods for extrapolation of episodic results for longer-term exposure time-scales. Along these lines, research was initiated in 1998 to acquire and apply Computational Fluid Dynamics (CFD) models, with the goal to simulate the pathway from the source of emissions to human exposures.

5.5 One Atmosphere

Bringing knowledge together (as a junction of streams, for the purpose of understanding) to form what is now commonly called one atmosphere modeling is an apt characterization of the period from 1990 to present. In truth, bringing knowledge together is how model development has always occurred. The decade, 1990 to 2000, was special because of the advancement in computer memory capacities and computational speed. This advancement allowed the blending together of more detailed and comprehensive descriptions of atmospheric physics and chemistry that heretofore had been forced by numerical capabilities to be treated either simplistically or separately. In anticipation of new computing capabilities and the possible consequences, Congress passed the High Performance Computing Act in 1991 (Public Law 102-194), which authorized a 5-year program with initial focus on high-speed parallel computing and networking. The additional funds, resulting from participating in this initiative, made it possible for the EPA to blend together research results from oxidant modeling with ROM and acid precipitation modeling with RADM, and also to develop and incorporate a

[26] For status of these activities, consult: http://www.epa.gov/ttn/atw/nata/.

pragmatic and innovative simulation of aerosol dynamics and chemistry all under one modeling framework. This allowed assessment of pollutant impacts as a complex mixture, rather than treating each pollutants effect independently.

The High Performance Computing and Communication Program (HPCC) is part of a larger Federal program sanctioned under the High Performance Computing Act of 1991 (Public Law 102-194) and coordinated through the Committee on Information and Communications of the National Science and Technology Council. The major program goals are: 1) build advanced capabilities to address multi-pollutant and multimedia issues; 2) adapt environmental management to high performance computing and communications environments; and 3) provide a modeling and decision support environment that is easy to use and responsive to environmental problem solving needs of key State, Federal and Industrial users.

There was nearly a 5-year lag from 1991 before shifts can be seen that directly related to High Performance Computing Act funds. The design of the Models-3 Framework and CMAQ modeling system was drafted in 1993 and 1994, and workshops were held in August and September of 1994 to assist in finalizing the concept designs. From these deliberations, the minimum acceptable functionality and the targeted capabilities were formalized for the Initial Operating Version of Models-3/CMAQ (Byun et al., 1995a; Dennis et al., 1996). The Models-3 Framework paradigm was designed to have multiple processing layers, which would allow rapid updates and development without having to redesign the entire framework (Byun et al., 1995b).

The Models-3 Framework was designed to be the interface that assists the user in operating the modeling systems. It included various components (e.g., to define the pathways to files; to manage updates and modifications to the source code; to define the domain, coordinate system, and science assumptions of an application; to manage the emissions processing).

The Community Multi-scale Air Quality (CMAQ) modeling system was designed to be a third-generation model that could simultaneously simulate multiple air quality issues, including tropospheric ozone, fine particles, toxics, acid and nutrient deposition, and visibility degradation. The initial science capabilities of CMAQ were derived from ROM, RADM and RPM. Unique to CMAQ is its multi-scale capabilities so that separate models are not needed for urban and regional scale air quality modeling. To implement multi-scale capabilities in CMAQ, several issues had to be addressed, such as scalable atmospheric dynamics and generalized coordinates that depend on the desired model resolution. The meteorological model had to be capable of simulating either hydrostatic conditions for large regional scales, where the atmosphere is assumed to have a balance of vertical pressure and gravitational forces with no net vertical acceleration on larger scales, or non-hydrostatic conditions for smaller scales such as urban environs where hydrostatic balance cannot be assumed. Because CMAQ is designed to handle scale-dependent meteorological formulations, CMAQ's governing equations are

expressed in a generalized coordinate system. This approach ensures consistency between CMAQ and the meteorological modeling system. The generalized coordinate system determines the necessary grid and coordinate transformations, and it can accommodate various vertical coordinates and map projections. By making CMAQ a modeling system that addresses multiple pollutants and different spatial scales, CMAQ has a "one atmosphere" perspective.

One of the project goal was to deliver for public testing a beta-test version of the Initial Operating Version (IOV) by the summer of 1997. To accomplish this goal, a variety of modules had to be tested and incorporated into Models-3/CMAQ. The Aerosol and Visibility Module was developed from the RPM and the deciview was added as a visibility metric (Pitchford and Malm, 1994). An advanced module was developed for specifying the photodissociation rates (Roselle et al., 1996) with the rates computed hourly for a gridded domain using clear-sky radiation rates, modeled temperatures, pressure, cloud fields, albedo and total ozone column data. A plume-in-grid module was developed to provide a more realistic treatment of the subgrid-scale physical and chemical processes affecting emissions from major elevated point sources (Godowitch et al., 1995). Further investigations were completed that demonstrated that the aggregation method, developed for the NAPAP RADM assessment, was capable of estimating long-term impacts from a sample of shorter episodes (Eder and LeDuc, 1996a, b; Cohn et al., 1999), and thus, solidifying the basis for incorporating the method into the Models-3 Framework. Initially, the Geocoded Emission Modeling and Projection (GEMAP) system was used as a basis for developing a flexible emissions processing system (Wilkinson et al., 1994). GEMAP was enhanced to include BEIS2 (Geron et al., 1994, 1995) for estimation of biogenic emissions, the RADM chemical speciation mechanism, the Carbon Bond IV mechanism, and the University of California's State Air Pollution Research Center (SAPRC) mechanisms (Lurmann, 1991). When further enhancements were made to GEMAP in 1997, to include the EPA Mobile 5a mobile-source emission processor, and to include links to the MM5 meteorological model, the new emissions processing system came to be called the Models-3 Emission Processing and Projection System (MEPPS).

In 1998, work began to replace MEPPS with the Sparse Matrix Operator Kernel Emissions (SMOKE©, developed by MCNC-North Carolina Supercomputing Center), which employs a matrix approach to complete the repetitive computations involved in producing very large emission databases. It is anticipated that SMOKE© will be at least an order of magnitude faster than MEPPS. In June 1998, Models-3/CMAQ version 2 was made available. To save expenses, the initial design of the Framework involved managing and sharing of information between several third-party programs. It became a resource intensive activity to adapt the Framework to various workstation environments, and to revise the Framework to be compatible with format changes resulting from upgrades in the third-party software. Since resources were never allocated to convert the Framework to one operating system, the Framework operations were reduced and simplified in each of the successive releases of the new versions of Model-3/CMAQ. The release of

Models-3/CMAQ version 3 in June of 1999 included Y2K updates to the support software, except for MM5. The release of Models-3/CMAQ version 4, in June 2000, simplified the installation procedures, and improvements were made to provide support for software updates and user help via the Models-3 web site[27].

6 Current Issues and Trends in Model Development

The purpose of the preceding discussions was to provide a historical review of the development of regulatory air quality models within the United States as viewed from the EPA, with a focus on the NOAA ARL scientists who provided EPA meteorological and air quality modeling support.

The Atomic Energy Act of 1954 established the requirement for Weather Bureau Field Offices to provide technical services in environmental impact assessment to Federal agencies. President Nixon's Reorganization Plan Number 4 in 1970 established the NOAA, and established a basis for NOAA to provide EPA support in the development of air quality models. In the late 1960s and early 1970s, the urban-scale long-term air quality models provided a proof of concept that air quality models could be used by the states to develop state implementation plans (SIPs) to relate the control of emissions with estimated air quality impacts, and thereby provide a rationale for how the States could achieve the NAAQS set by the EPA. The early models evolved from the Gaussian plume model that was developed in England during the period of 1916 to the early 1930s to investigate the effects of gas warfare. The special needs of characterizing air quality impacts from industrial stack emissions stimulated research into buoyant plume rise and convective dispersion processes. Wind tunnel investigations provided a basis for investigating building wake effects on atmospheric dispersion. As these investigations evolved, Gaussian plume models were provided with new capabilities that allowed their use to satisfy the Clean Air Act Amendments of 1977, accepting usage of air quality models as a means for demonstrating compliance for the development of State Implementation Plans and for permits required by the New Source Review program. By the late 1970s, Gaussian plume models had gained widespread use in regulatory assessments, and during the late 1990s, efforts were underway to extend the usefulness of these models for more complex transport situations through the use of puff dispersion models. During the early 1970s, tropospheric photochemical ozone models were being developed to characterize ozone impacts within a city. These early grid models, struggling with poorly understood chemical kinetics and uncertain emission inventories, were severely limited by the memory capabilities and computation speed of computer technology. As field and laboratory studies began to resolve the uncertainties in the physical processes, it became increasingly clear that the scale of the problem was much larger than the domain of a city or even a state. The Clean Air Act Amendments of 1990 required the use tropospheric chemistry air quality models

[27] For current status of Models-3/CMAQ consult: http://www.epa.gov/asmdnerl/CMAQ/index.html.

for demonstrating a plan for attainment of the ozone NAAQS. By the 1990s, advances in scientific understanding of ozone formation, acid deposition and aerosols coupled with advances in computer science provided a basis for the development of second and third generation (one atmosphere) regional-scale chemistry-aerosol grid models for use in regulatory assessments. It is seen that the Clean Air Act Amendments of 1977 and 1990 formalized the acceptance and use of air quality models in regulatory assessments. In the 1990s, EPA placed increased emphasis towards characterizing human exposures to toxics, which may in some instances require simulation of air chemistry. Human exposure models couple the time history of pollutant species within microenvironments with human activity patterns. Such models have increased interest in the development of air quality models that can address neighborhood scale impacts within comprehensive grid models.

In developing this review, three issues seem to resurface more than once, and appear to deserve special comment, namely: 1) a trend to require a complete documentation of the assumptions made (transparency) and a formal exploration of the consequences of these assumptions in air quality assessments, 2) a need for the use of standardized methods in development of emission inventories, and 3) a need for development and use of science-based model performance standards.

6.1 Transparency and Consequences of Assumptions

The recommendations by the National Academy of Sciences Committee on the Institutional Means for Assessment of Risks to Public Health (National Research Council, 1983) planted a seed within the U.S. Congress and within the EPA regulatory culture that formalized uncertainty assessments to promote acceptance of regulatory policy decisions. Prompted by the significant scientific uncertainties in the health effects and in the available characterizations for long range transformation-transport of sulfate, Congress passed the Acid Precipitation Act of 1980 (Public Law 96-294). This directed the United States Government to conduct a ten-year assessment to determine the causes and consequences of acid precipitation, and to develop options for reducing known effects. The National Acid Precipitation Assessment Program (NAPAP) was created in response to this congressional charge. The NAPAP assessment of 1990 validated the effort that went into the development of the Regional Acid Deposition Model (RADM).

One of the recommendations, resulting from the National Research Council's (1991) review of the state of knowledge of the physical and chemical sciences relevant to the characterization of tropospheric ozone, was that periodic reviews of this kind were needed. In response to this recommendation, the North American Research Strategy for Tropospheric Ozone (NARSTO)[28] was formed with over 70

[28] http://eosweb.larc.nasa.gov/GUIDE/campaign_documents/base_narsto_project.html.

participating organizations from Canada, Mexico and the United States. During their deliberations, it was decided to broaden the purview to include fine particles. Twenty-four critical reviews of the science disciplines involved in characterizing tropospheric ozone were prepared as part of the NARSTO assessment, of which seventeen were published in Volume 34 of *Atmospheric Environment*. The focus of the assessment was on the state of science knowledge and possible regulatory implications. It was concluded that the main science advances from 1990 to 2000 mostly confirmed hypotheses and results reported earlier. With each assessment, there can be seen an increased attention to attempt to make transparent the assumptions, not only of the science being reviewed, but of the assessment process itself, to critically review each assumption, and to attempt to assess the known uncertainties on conclusions reached.

The goals, to state the scientific assumptions and to explore the consequent uncertainties on conclusions reached, have been expressed in the guidelines for exposure assessment (U.S. Environmental Protection Agency, 1992, http://www.epa.gov/ncea/exposure.htm, and 1998, http://www.epa.gov/ncea/ecorsk.htm), which describe the general concepts of human and ecological exposure assessments, including recommendations on the presentation of results and the characterization of uncertainty. These guidelines recommend that assumptions be explicitly stated, and the uncertainties associated with these assumptions be discussed as well as the possible implications on conclusions reached. In this context, it is worth noting that the 1990 CAAA called for residual risk assessments to be conducted to assess whether further reductions are needed in order to mitigate exposures to toxic emissions. The large NRC and NARSTO assessments, the EPA guidelines and the 1990 CAAA all support a trend for creation and use of large science committees to provide independent assessments of the science progress with a focus on the implications for regulatory decisions. It is reasonable to anticipate that future pollution mitigation programs will increasingly require public acceptance, and to gain this support the regulatory community will increasingly call upon formal assessments to make transparent the assumptions made and consequent uncertainties. The strength of these assessments is that they are considered to be unrehearsed, unbiased, and have no hidden agenda. To date, the integrity of these assessments has successfully relied on the integrity of the individuals involved. As the climate surrounding the decision making process becomes more contentious, there may be a call to define formal assessment procedures. Developing standardized procedures that foster making the science assumptions and decision assumptions transparent, and for characterizing the consequences of these assumptions will be a trend to look for in the future.

6.2 Emission Inventories

Assembling and checking the quality of air quality models' emission inventory has always been recognized as a most difficult task. Seinfeld (1988) described the emission inventories for then typical urban-scale ozone air quality model applications as being immense databases that might contain on the order of 500 to several thousand individual point sources and up to several hundred area source categories. Biogenic and vegetative emissions of volatile organics have historically been largely neglected. He characterized the techniques used to quantify uncertainties relying upon the judgment of the inventory specialist. The National Research Council's (1991a) review of ozone modeling expressed concerns because of the underestimation of anthropogenic VOC emissions that 1) mandated emission controls in past years were a smaller fraction of the total than originally anticipated, and thus, limited the effectiveness of these controls, and 2) planned control strategies for future years will likely require fundamental revisions. The critical review by Placet et al. (2000) concluded that the standard emissions factors (U.S. Environmental Protection Agency, 1985b) were representative of older industries constructed prior to 1970. The three main steps (speciation, temporal scaling, spatial allocation), needed to convert the emission inventories for use by air quality models, were seen to be crude and uncertain. Projecting the consequences of these uncertainties on conclusions reached in emission control strategies was seen as problematic and difficult. The investigations by Hanna et al. (1998, 2001) estimated the uncertainty distribution for the domain-wide maximum ozone concentration was close to log-normal, with 95% of the values within a factor of ±1.6 from the median, with uncertainties in anthropogenic VOC emissions from area sources having the most influence.

Although one might call for extensive programs to assess and reduce emission inventory uncertainties, the reality of the situation is that most of the problems are well identified and progress is being made within the limits imposed by available resources. The uncertainties in those sources characterized as area sources (mobile, small businesses, biogenics) are recognized and efforts are underway to develop improvements. Results from the Southern Oxidant Study (SOS) (Chameides and Cowling, 1995) indicate that oxides of nitrogen emissions from Tennessee, for example may be comparable with those of the coal-fired power plants in the state (Williams et al., 1992; Valente and Thornton, 1993). The 1992 SOS tunnel studies suggest that improvements have been made in our ability to characterize mobile emissions (Pierson et al., 1996). Problems in the characterization of biogenic emissions are being traced to uncertainties in characterizing land use, so efforts are underway to develop improved land use maps. Thus, of all the actions that have been suggested, one of the more pragmatic and cost effective suggestion appears to be the proposal to promote the use of EPA's Emission Inventory Improvement Program (EIIP, 1996). This is a set of standard procedures that provide preferred and alternative methods for emissions estimation; if employed, it would promote consistency. Improvements would still be needed in the estimates of emission factors, and assumptions used (e.g.,

effectiveness of control, activity levels, growth rates). However, promoting use of standardized procedures that foster consistency in emission inventory development will be a trend to look for in the future.

6.3 Model Performance Standards

One of the luxuries of developing new characterizations of dispersion of a relatively inert pollutant from an isolated source is that tracer field data studies can be used to evaluate the model performance. This is not the case for tropospheric models that characterize chemical processes and aerosol processes, which means that uncertainties in emission characterization becomes a part of the problem in evaluating the performance of these models. In spite of these differences and many attempts that have been made[21], consensus standards have yet to be devised for assessing air quality model performance. Turner (1979) noted that there were no recognized model performance standards (metersticks). Dennis and Downton (1984), Seinfeld (1988), and Russell and Dennis (2000) all have noted that evaluations conducted have not tested the suitability of tropospheric air chemistry models to simulate changes in air quality that involve substantial changes in emissions. Russell and Dennis (2000) further note that simple comparisons of observed and simulated ozone values are insufficient to determine whether the chemical processes were properly characterized.

In 1996, an effort was initiated within the American Society for Testing and Materials (ASTM, http://www.astm.org) to foster the development of standard methods for evaluating the performance of air quality models. The first product of this effort was a Standard Guide for the Statistical Evaluation of Atmospheric Dispersion Model Performance (D 6589). Within the annex of this guide, an example procedure was defined that would allow testing of dispersion models in their ability to characterize the average maximum concentration as a function of distance downwind from an isolated source whose emissions can be treated as being relatively inert. This procedure also provides an objective statistical test to define when differences in performance are significant. The plans are to create a series of formal ASTM Test Methods ("metersticks" as called by Turner in 1979) that can be cited by regulatory agencies as approved methods for characterizing model performance. The initial work will focus on dispersion models, and then as experience is gained, expand the purview to include models that characterize atmospheric chemistry and aerosol formation processes. Developing standard "metersticks" of model performance within ASTM has the potential for having far reaching effects, as it would provide an international basis for comparing model development efforts and establishing international acceptance standards for air quality models for use in legal proceedings and regulatory decision making.

Acknowledgments

This work was funded in part by the United States Environmental Protection Agency through an Interagency Agreement (139-384-83) with the National Oceanic and Atmospheric Administration.

It has been subjected to Agency review and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

References

Alm, L.R, 2000. *Crossing Borders, Crossing Boundaries: the Role of Scientists in the U.S. Acid Rain Debate*. Praeger Publishers, 147 pages.

Anthes, R.A., Seaman, N., Sobel, J., and Warner, T., 1974. *The development of mesoscale models suitable for air pollution studies*. Select Research Group in Air Pollution Meteorology, 2nd Annual Report: Vol. I, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, pp. 6-233.

Anthes, R.A., and Warner, T., 1978. Development of hydrodynamic models suitable for air pollution and other mesometeorological studies. *Monthly Weather Review*. 106:1045-1078.

Anthes, R.A., Hsle, E.Y., and Kuo, Y.H., 1987. *Description of the Penn State/NCAR Mesoscale Model version 4 (MM4)*, NCAR Technical Note, NCAR/TN-282+STR, National Center for Atmospheric Research, Boulder, CO, 66 pages.

Arnts, A., and Meeks, S., 1981. Biogenic hydrocarbon contribution to the ambient air of selected areas. *Atmosphere Environment*, 15(9):1643-1651.

Barad, M.L., and Haugen, D.A., 1959. A preliminary evaluation of Sutton's hypothesis for diffusion from a continuous point source. *Journal of Meteorology*. Vol. 16(1):12-20.

Benkley, C.W., and Bass, A., 1979. *User's guide to MESOPUFF (Mesoscale Puff) Model*. Environmental Research and Technology, Inc., Concord, MA.

Bennet, M., and Hunter, G.C., 1997. some comparisons of lidar estimates of peak ground-level concentrations with the predictions of UK-ADMS. *Atmospheric Environment*. 31(3):429-439.

Biller, W.R., Feagans, T.B., Johnson, T.R., Duggan, G.M., Paul, R.A., McCurdy, T., and Thomas, H.C., 1981. A general model for estimating exposure associated with alternative NAAQS. Paper No. 81-18.4. In *Proceedings of the 74th Annual Meeting of the Air Pollution Control Association*, June 21-26, 1981, Philadelphia, PA, 25 pages.

Binkowski, F.S., and Ching, J.K.S., 1996. Modeling the particulate mass and visibility using the EPA Regional Particulate Model. Preprints of the *9th Joint Conference on Applications of Air Pollution Meteorology with A&WMA*, January 28-February 2, 1996, Atlanta, GE, American Meteorological Society, Boston, MA, pages 565-569.

Binkowski, F.S., and Shankar, U., 1994. Development of an algorithm for the interaction of a distribution of aerosol particles with cloud water for use *in a three-dimensional Eulerian air quality model*. *Abstracts, 4th* International Aerosol Conference, August 29-September 2, 1994, Los Angeles, CA, Richard C. Flagan (Ed.), American Association for Aerosol Research, Cincinnati, OH, page 220.

Binkwoski, F.S., and Shankar, U., 1995. The Regional Particulate Matter Model, 1. Model description and preliminary results. *Journal of Geophysical Research*. 100(D12):26,191-26,209.

Binkowski, F.S., Chang, J.S., Dennis, R.L., Reynolds, S.J., Samson, P.J., and Shannon, J.D., 1990. Regional acid deposition modeling. Acidic Deposition. State of Science and Technology Report 3. *Volume 1, Emissions, Atmospheric Processes and Deposition, National Acid Precipitation Assessment Program*, 722 Jackson Place, NW, Washington DC.

Briggs, G., 1965. *A Plume Model Compared With Observations*. Paper 65-44 presented at Annual Meeting of the Air Pollution Control Association, June 20-24, 1965, Toronto, Ontario, 23 pages.

Briggs, G.A., 1969. *Plume Rise*. TID-25075. U.S. Atomic Energy Commission, Available from the National Technical Information Service, U.S. Department of commerce, Springfield, VA 22151, 85 pages.

Briggs, G.A., 1975. Plume Rise Predictions. *Lectures on Air Pollution and Environmental Impact Analyses*, D. A. Haugen, Ed., American Meteorological Society, Boston, pages 59-111.

Briggs, G.A., 1989. Field measurement of vertical diffusion in convective conditions. Preprints, *6th Joint Conference on Applications of Air Pollution Meteorology*, January 30-February 3, 1989. Anaheim, CA, American Meteorological Society, Boston, MA, pages 167-170.

Briggs, G.A., 1993a. Final results of the CONDORS convective diffusion experiment. *Boundary Layer Meteorology*, Dordrecht, Then Netherlands, 62(1-4):315-328.

Briggs, G.A., 1993b. Plume dispersion in the convective boundary layer, Part II, Analyses of CONDORS field experiment data. *Journal of Applied Meteorology*, American Meteorological Society, Boston, MA, 32(8):1388-1425.

Bullock, O.R. Jr., 1990. The effect of size-dependent dry deposition velocities in an Eulerian regional-scale particulate model. Preprints, *18th International Technical Meeting of NATO/CCMS on Air Pollution Modeling and its Application*, Volume 1, May 13-17, 1990, University of British Columbia, Vancouver, BC, Canada. NATO/CCMS, Brussels, Belgium, pages 145-152.

Bullock, O.R. Jr., Roselle, S.J., and Heilman, W.E., 1989. *Development and preliminary testing of a first-generation regional aerosol model*. Status report, Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC 109 pages.

Byun, D.W., Hanna, A., Coats, C., and Hwang, D., 1995b. Models-3 air quality model prototype science and computational concept development. In Regional Photochemical Measurement and Modeling Studies. TR-24. *Transactions of an International Specialty Conference*, San Diego, CA, November 8-12, 1993. Air & Waste Management Association, Pittsburgh, pages 197-212.

Byun, D.W., Coats, C.J., Hwang, D., Fine, S., Odman, T., Hanna, A., and Galluppi, K.J., 1995a. Prototyping and implementation of the multi-scale air quality models for high performance computing. In High Performance Computing Symposium 1995; Grand Challenges in Computer Simulation. Proceedings of the *1995 Simulation Multiconference, Phoenix, AZ, April 9-12, 1995*. Adrian M., Tentner (Ed.), The Society for Computer Simulation, San Diego, CA, pages 527-532.

Calder, K.L., 1969. *A Narrow Plume Simplification For Multiple Urban Source Models*. (Unpublished note from K.L. Calder to F.A. Gifford), 11 pages.

Calder, K.L., 1971. A climatological model for multiple source urban air pollution. Paper presented at First Meeting of the NATO/CCMS Panel on Modeling. Paper published in Appendix D of *User's Guide for the Climatological Dispersion Model*. EPA-R4-73-024. Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, pages 73-105.

Calder, K.L., 1977. Multiple-source plume models of urban air pollution - their general structure. *Atmospheric Environment*. Vol. 11, pages 403-414.

Carruthers, D.J., Holroyd, R.J., Hunt, J.C.R., Weng, W.S., Robins, A.G., Apsley, D.D., Thomson, D.J., and Smith, F.B., 1994. UK-ADMS: a new approach to modelling dispersion in the earth's atmospheric boundary layer. *Journal of Wind Engineering*. (52):139-153.

Carter, W.P.L., Lurmann, F.W., Atkinson, R., and Lloyd, A.C, 1986. Development and testing of a surrogate species chemical reaction mechanism, Volumes I and II. EPA-600/3-86-031, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

Chameides, W.L., and Cowling, E.B., 1995. The state of the Southern Oxidants Study. Policy Relevant Findings in Ozone Pollution Research, 1988-1994. North Carolina State University, Raleigh, NC.

Chameides, W., Lindsay, R., Richardson, J., and Klang, C., 1988. The role of biogenic hydrocarbons in urban photochemical smog. Atlanta as a case study. Science, 241:1473-1475.

Chang, J.S., Middleton, P.B., Stockwell, W.R., Walcek, C.J., Pleim, J.E., Lansford, H.H., Binkowski, F.S., Seaman, N.L., McHenry, J.N., Madronich, S., Samson, P.J., and Hass, H., 1990. The regional acid deposition model and engineering model. Acidic Deposition. State of Science and Technology Report 4. Volume 1, Emissions, Atmospheric Processes and Deposition, National Acid Precipitation Assessment Program, 722 Jackson Place, NW, Washington DC.

Ching, J.K.S., and Doll, D.C., 1981. Temporal variation of ground hear flux for soil and concrete using net radiation data. *Proceedings of the 5th Symposium on Turbulence, Diffusion and Air Pollution*, March 9-13, Atlanta, GA., American Meteorological Society, Boston, MA, pages 171-172.

Clark, T.L., 1992. Atmospheric deposition of toxic metals to Lake Michigan: Preliminary annual model calculations. Proceedings of the 1992 U.S. EPA/A&WMA International Symposium on Measurement of Toxic Air Pollutants. Durham, NC, May 1992. U.S. Environmental Protection Agency, Research Triangle Park, NC, and Air & Waste Management Association, Pittsburgh, PA, pages 681-686.

Clark, T.L., Blakley, P., and Mapp, G., 1992. Model calculations of the annual atmospheric deposition of toxic metals to Lake Michigan. Abstracts Book from the Air & Waste Management Association 86th Annual Meeting, Kansas City, Missouri, June 21-26, 1992. Air & Waste Management Association, Pittsburgh, PA, 92-84.17.

Clarke, J.R., Ching, J.K.S., and Godowitch, J.M., 1981. Spatial characteristics of surface boundary layer turbulence in an urban area. *Proceedings of the 5th Symposium on Turbulence, Diffusion and Air Pollution*, March 9-13, Atlanta, GA., American Meteorological Society, Boston, MA, pages 167-168.

Cohn, R., Eder, B.K., and LeDuc, S.K., 1999. An aggregation and episode selection scheme designed to support Models-3 CMAQ. In *Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) modeling system*. Chapter 17, D.W. Byun, and J.K.S. Ching (Eds.). EPA/600/R/99/030. National Exposure Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC, pages 1-66.

Cowling, E.B., 1992. The performance and legacy of NAPA. *Ecological Applications*, 2(2):111-116.

Cramer, H.E., 1957. A practical method for estimating the dispersal of atmospheric contaminants. *Proceeding of 1st National Conference on Applied Meteorology*. American Meteorological Society. Hartford, CT. pages C-33 through C-55.

Crutzen, P.J., 1973. A discussion of the chemistry of some minor constituents I the stratosphere and troposphere, *Pure Appli. Geophys*., 106-108:1385-1399.

Crutzen, P.J., 1974. Photochemical reactions initiated by and influencing ozone in unpolluted tropospheric air. *Tellus*, 26:47-57.

DeMarrais, G.A., 1974. *A History of Air Pollution Meteorology Through 1969.* NOAA Technical Memorandum ERL ARL-74. Air Resource Laboratory, Silver Spring, MD. 78 pages.

Dennis, R.G., 1997. Using the Regional Acid Deposition Model to determine the nitrogen deposition airshed of the Chesapeake Bay watershed. In *Atmospheric Deposition of Contaminants to the Great Lakes and Coastal Waters*. J.E. Baker (Ed.). STAC Press, Pensacola, FL, pages 393-413.

Dennis, R.,L., and Downton, M.W., 1984. Evaluation of urban photochemical models for regulatory use. *Atmospheric Environment*. 18:2055-2069.

Dennis, R.L., Byun, D.W., Novak, J.H., Galluppi, K.J., Coats, C.J., and Vouk, M.A., 1996. The next generation of integrated air quality modeling: EPA's Models-3. *Atmospheric Environment*. 30:1925-1938.

Dennis, R.G., Barchet, W.R., Clark, T.L, McHenry, J.N, and Reynolds, S.D. , 1990a. Evaluation of regional acidic deposition models. Acidic Deposition. State of Science and Technology Report 5. *Volume 1, Emissions, Atmospheric Processes and Deposition, National Acid Precipitation Assessment Program*, 722 Jackson Place, NW, Washington, DC.

Dennis, R.G., Binkowski, F., Clark, T., McHenry, J., Reynolds, S., and Seilkop, S., 1990b. Selected application of the Regional Acid Deposition Model and Engineering Model. Acidic Deposition. State of Science and Technology Appendix 5F. *Volume 1, Emissions, Atmospheric Processes and Deposition*, National Acid Precipitation Assessment Program, 722 Jackson Place, NW, Washington, DC.

Dewert, R.G., and Stewart, H.N.M., 1973. Elevated ozone levels in the air of Central London, *Nature*. (241):342-343.

Dodge, M.C., 1977. Combined use of modeling techniques and smog chamber data to derive ozone - precursor relationships. Proceedings of the *International Conference on Photochemical Oxidant Pollution and Its Control*, EPA-600/3-77-001b. U.S. Environmental Protection Agency, Research Triangle Park, NC 27711. pages 881-889.

Donigian, A.S., Jr., Bicknell, B.R., Patwardhan, A.S., Linker, L.C., Alegre, D.Y., Chang, C.H., and Reynolds, R., 1991. *Watershed Model Application to Calculate Bay Nutrient Loads: Phase II Findings and Recommendation*. U.S. Environmental Protection Agency, Chesapeake Bay Program, Annapolis, MD.

Duan, N., 1981. Micro-Environmental Types: A Model for Human Exposure to Air Pollution. *SIMS Technical Report No. 47*, Stanford California Department of Statistics, Stanford University, 24 pages.

Duan, N., 1989. Estimation of microenvironmental concentration distributions using integrated exposure measurements. Proceedings of the *Research Planning Conference on Human Activity Patterns*. T.H. Stars (Ed.). EPA/600/4-89/004. Office of Research and Development, U.S. Environmental Protection Agency, Las Vegas, NV, pages 15-1 to 15-14.

Eberhard, W.L., Moninger, W.R., Briggs, G.A., 1988. Plume dispersion in the convective boundary layer, Part I, CONDORS field experiment and example measurements. *Journal of Applied Meteorology*, American Meteorological /Society, Boston, MA, 27(5):599-616.

Eder, B.K., and LeDuc, S.K., 1996a. Aggregation of selected RADM simulations to estimate annual ambient air concentrations of fine particulate matter. Preprints, *9th Joint Conference on the Applications of Air Pollution Meteorology with A&WMA*. January 28-February 2, 1996, Atlanta, Georgia. American Meteorological Society, Boston, MA, pages 390-392.

Eder, B.K., and LeDuc, S.K., 1996b. Can selected RADM simulations be aggregated to estimate annual concentrations of fine particulate matter? In *Measurement of Toxic and Related Air Pollutants. VIP-64. Proceedings of an International Specialty Conference*, Research Triangle Park, NC, May 7-10, 1996, U.S. Environmental Protection Agency, RTP, NC and Air & Waste Management Association, Pittsburgh, PA, pages 732-739.

Eder, B.K., Coventry, D.H., Clark, T.L., and Bollinger, C.E., 1986. *RELMAP: A Regional Lagrangian Model of Air Pollution user's guide*. EPA/600/8-86/013. Atmospheric Sciences Research Laboratory, Research Triangle Park, NC, 137 pages.

Electric Power Research Institute 1984. *Regional air quality model assessment and evaluation*. EA-36-71, Research Project 1630-21, Electric Power Research Institute, Palo Alto, CA.

Eliassen, A., 1978. The OECD study on long range transport of air pollutants, long range transport modelling. *Atmospheric Environment*. Vol. 12:479-487.

Eliassen, A., 1980. A review of long-range transport modeling. *Journal of Applied Meteorology*. Vol. 19(3):231-240.

Eliassen, A., Hov, Ø., Isaksen, I.S.A., Saltbones, J., and Stordal, F., 1982. A Lagrangian long-range transport model with atmospheric boundary layer chemistry. *Journal of Applied Meteorology*. Vol. 21:1645-1661.

Emissions Inventory Improvement Program, 1997. EIPP Technical Report Series, Volumes I through X, available from: http://www.epa.gov/ttn/chief/eiip/techreport/index.html.

Eschenroeder, A.Q., and Martinez, J.R., 1971. *Concepts and application of photochemical smog models*. Technical Memo 151B. General Research Corporation, Santa Barbara, CA.

Eschenroeder, A.Q., Martinez, J.R., and Nordsieck, R.A., 1972. *Evaluation of a diffusion model for photochemical smog simulation*. CR-1-273. General Research Corporation, Santa Barbara, CA.

Federal Register, 1980. Reconsideration of the use of Pasquill-Gifford dispersion coefficients for stability class A in setting emission limitations for four Ohio power plants; petitions for reconsideration. *45 FR 74041*, pages 74041-74047.

Ferber, G.J., Heffter, J.L., Draxler, R.R., Lagomarsino, R.J., Thomas, F.L., Dietz, R.N., Benkovitz, C.M., 1986. Cross-Appalachian Tracer Experiment (CAPTEX 83) Final Report. *NOAA Technical Memorandum ERL ARL-142*, Air Resources Laboratory, Silver Spring, MD, 60 pages.

Fisher, B.E.A., 1983. A review of the processes and models of long-range transport of air pollutants. *Atmospheric Environment*. Vol. 17(10):1865-1880.

Fox, D.G., 1981): Judging air quality model performance. *Bulletin of the American Meteorological Society*. (62):599-609.

Fox, D.G., 1984. Uncertainly in air quality modeling. *Bulletin of the American Meteorological Society*. (65):27-36.

Frenkiel, F.N., 1957. Atmospheric pollution in growing communities. *Publication 4276. Smithsonian Report for 1956,* Smithsonian Institution, Washington, DC, pages 269-299.

Friedlander, S.K., and Seinfeld, J.H., 1969. A dynamic model of photochemical smog. *Environmental Science & Technology*. 3(11):1175-1181.

Gelbard, F., 1984. MAEROS. *Aerosol Science and Technology*. 3:117-118.

Gelbard, F., and Seinfeld, J.R., 1980. Simulation of multicomponent aerosol dynamics. Journal Colloid Interface Science. 78:485-501.

Geron, C.D., Guenther, A.B., and Pierce, T.E., 1994. An improved model for estimating emission of volatile organic compounds from forests in the eastern United States. *Journal of Geophysical Research*. 99(D6):12,773-12,791.

Geron, C.D., Pierce, T.E., and Guenther, A.B., 1995. Reassessment of biogenic volatile organic compound emissions in the Atlanta area. *Atmospheric Environment.* 29:1569-1578.

Gery, M.W., Whitten, G.Z., and Killus, J.P., 1988. *Development and Testing of the CBM-IV for urban and regional modeling.* EPA/600/3-88/012, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711l.

Gifford, F.A., Jr. 1961. Uses of routine meteorological observations for estimating atmospheric dispersion. *Nuclear Safety*. Vol. 2(4):47-51.

Gifford, F.A., 1976. Turbulent diffusion-typing schemes: a review. *Nuclear Safety*. Vol 17(1):68-86.

Gifford, F.A. and Hanna, S.R., 1970. Urban air pollution modelling. *Proceedings of the Second International Clean Air Congress*. Paper ME-320. Washington, D.C., pages 1146-1151.

Gifford, F.A. and Hanna, S.R., 1973. Modeling urban air pollution. *Atmospheric Environment*. (7):131-136.

Glen, W.G., and Zelenka, P.J., 1994. Trends in CO exposure for Denver using population based exposure model. Abstracts Book, 4th Conference of the International Society for Exposure Analysis, Research Triangle Park, North Carolina, September 18-21, 1994. International Society for Exposure Analysis, Host, School of Public Health, University of North Carolina at Chapel Hill, Chapel Hill, NC, page 146.

Godowitch, J.M., Ching, J.K.S., and Clarke, J.F., 1979. Dissipation of the nocturnal inversion layer at an urban and rural site in St. Louis, MO. *Proceedings of the 4th Symposium on Turbulence, Diffusion and Air Pollution.* January 15-18, Reno, NV, American Meteorological Society, Boston, MA, pages 416-420.

Godowitch, J.M., Ching, J.K.S., and Clarke, J.F., 1981. Urban/rural and temporal variations in PBL turbulence parameters and length scales over St. Louis, MO. *Proceedings of the 5th Symposium on Turbulence, Diffusion and Air Pollution.* Atlanta, GA, American Meteorological Society, Boston, MA, pages 171-172.

Godowitch, J.M., Ching, J.K.S., and Gillani, N.V., 1995. A treatment for Lagrangian transport and diffusion of subgrid scale plumes in an Eulerian grid framework. Preprints, *11th Symposium on Boundary Layers and Turbulence*. March 27-31, 1995, Charlotte, NC. American Meteorological Society, Boston, pages 86-89.

Grell, G.A., Dudhia, J., and Stauffer, D.R., 1994. A Description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5). *NCAR Technical Note, NCAR/TN-398+STR*, National Center for Atmospheric Research, Boulder, CO, 138 pages.

Gryning, S.E., Holtslag, A.A.M., Irwin, J.S, Sivertsen, B., 1987. Applied dispersion modelling based on meteorological scaling parameters. *Atmospheric Environment*. Vol. 21(1):79-89.

Guicheri, R., and van Dop, H., 1977. Photochemical production of ozone in Western Europe (1971-1975) and its relation to meteorology. *Atmospheric Environment.* (11):145-155.

Haagen-Smit, A.J., 1950. The air pollution problem in Los Angeles. *Engineering Science*. Vol. 14:7-14.

Haagen-Smit, A.J., 1952. Chemistry and physiology of Los Angeles smog. *Industrial Engineering Chemistry*. Vol. 44:1342

Haagen-Smit, A.J. and Fox, M.M., 1955. *SAE Transactions*, Vol. 63:575.

Hamming, W.J., MacPhee, R.D., and Taylor, J.R., 1960): Contaminant concentrations in the atmosphere of Los Angeles County. *Journal of Air Pollution Control Association.* Vol. 10(1):7-16.

Hanna, S.R., and Chang, J.C., 1991. Revision of the hybrid plume dispersion (HPDM) for application to urban areas. H. van Dop and D.G. Steyn (Editors), *Air Pollution Modeling and its Application VIII*, Plenum Press, New York, NY, pages 751-758.

Hanna, S.R., and Paine, R.J., 1989. Hybrid plume dispersion model (HPDM) development and evaluation. *Journal of Applied Meteorology*. American Meteorological Society, Boston, MA, 28(3):206-224.

Hanna, S.R., Briggs, G.A., and Hosker, R.P., 1982. *Handbook on Atmospheric Diffusion*. Available as DE82002045 (DOE/TIC-11223) from the National Technical Information Service, U.S. Department of Commerce, Springfield, VA 22161, 108 pages.

Hanna, S.R.; Chang, J.C., and Fernau, M.E., 1998. Monte Carlo Estimates of Uncertainties in Predictions by a Photochemical Grid Model (UAM-IV) Due to Uncertainties in Input Variables, *Atmospheric Environment*, Vol. 32, pages 3617-3628.

Hanna, S.R., Zhigang, L., Christopher, F., Wheeler, N., Vukovich, J., Arunachalam, S., Fernau, M., and Hanse, D.A., 2001. Uncertainties in predicted ozone concentrations due to input uncertainties for the UAM-V photochemical grid model applied to the July 1995 OTAG domain. Atmospheric Environment. (35):891-903.

Hicks, B.B., Draxler, R.R., Albritton, D.L., Fehsenfeld, F.C., Dodge, M., Schwartz. S.E., Tanner, R.L., Hales, J.M., Meyers, T.P., and Vong, R.J., 1990. Atmospheric processes research and process model development. Acidic Deposition. State of Science and Technology Report 2. *Volume 1, Emissions, Atmospheric Processes and Deposition*, National Acid Precipitation Assessment Program, 722 Jackson Place, NW, Washington, DC.

Holland, J.Z., 1953. *A Meteorological Survey of the Oak Ridge Area: Final Report Covering the Period 1948-1952.* ORO-99, Technical Information Service, Atomic Energy Commission, Oak Ridge, TN, 584 pages.

Holzworth, G.C., 1964. Estimates of mean maximum mixing depths in the contiguous United States. *Monthly Weather Review*. Vol. 92(5):235-242.

Huber, A.H., and Snyder, W.H., 1976. Building wake effects on short stack effluents. Preprints of *Third Symposium on Atmospheric Diffusion and Air Quality*, American Meteorological Society, Boston, MA, pages 235-242.

Hudischewskyj, A.B., Saxena, P., and Seigneur, C., 1987. *Development of Computer Modules of Particulate Processes for Regional Particulate Model.* Contract No. 68-02-4076. Atmospheric Sciences Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, 284 pages.

Hunt, J.C.R., and Snyder 1980. Experiments on stably and neutrally stratified flow over a model three-dimensional hill. *Journal of Fluid Mech*. (98):671-704.

Hyde, W.L., 1952. *Atmospheric Radioactivity Following Nuclear Explosions*. Office of Naval Research, Technical Report ONRL-66-52, London, England.

Irwin, J.S. and Brown, T.M., 1985. A sensitivity analysis of the treatment of area sources by the Climatological Dispersion Model. *Journal of the Air Pollution Control Association.* Vol. 35(4):39-364.

Irwin, J.S., and Smith, M.E., 1984. Potentially useful additions to the rural model performance evaluation. *Bulletin of the American Meteorological Society*. (65):559-568.

Jasanoff, S., 1990. *The Fifth Branch, Science Adviser as Policymakers*. Harvard University Press, Cambridge, MA, 302 pages.

Johnson, T.R., 1983. *A Study of Personal Exposure to Carbon Monoxide in Denver, Colorado.* EPA 600/4-84-014. Office of Research and Development, Environmental Monitoring Systems Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC, 272 pages.

Johnson, T.R., 1992. *Estimation of Carbon Monoxide Exposures and Associated Carboxyhemoglobm Levels in Denver Residents Using a Probabilistic Version of NEM*. Report prepared by International Technology Air Quality Services, Contract 68-DO-0062, Work Assignment no. 1-4, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Johnson, T.R., and Paul, R.A., 1981. *The NAAQS Model (NEM) and Its Application to Particulate Matter*. Draft report prepared by PEDCo Environmental, Inc., Durham, NC., Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Johnson, T.R., and Paul, R.A., 1983. *The NAAQS Model (NEM) and Its Application to Carbon Monoxide*. EPA-450/5-83-003. Report prepared by PEDCo Environmental, Inc., Durham, NC., Contract 68-02-3390, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC., 197 pages

Johnson, T.R., and Paul, R.A., 1984. *The NAAQS Model (NEM) and Its Application to Nitrogen Dioxide*. Draft report prepared by PEDCo Environmental, Inc., Durham, NC., Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC. 71 pages.

Johnson, T.R., McCoy, M., and Capel, J.E., 1992. *Enhancements to the Hazardous Air Pollutant Exposure Model (HAPEM) as Applied to Mobile Source Pollutants*. Report prepared by IT Air Quality Services, Office of Air Quality Planning and Standards, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Kao, C.-Y. J., and Yamada, T., 1988. Use of the CAPTEX data for evaluations of a long-range transport numerical model with a four-dimensional assimilation technique. *Monthly Weather Review*, Vol. 116:293-306.

Karl, T.R., 1980. Study of the spatial variability of ozone and other pollutants at St. Louis, Missouri. *Atmospheric Environment*. 14(6):681-694.

Kingdon, J.W., 1995. A*genda, Alternatives and Public Policies*. 2nd edition, Harper Collins College Publishers, 254 pages.

Lackey, R.T., and Blair, R.L., 1997. Science, policy, and acid rain. *Renewable Resources Journal*. Vol. 15(1):9-13.

Lamb, B., Fuenther, A., Gay, D., and Westbery, H., 1987. A national inventory of biogenic hydrocarbon emissions. *Atmospheric Environment*, 21(8):1695-1705.

Lamb, R.G., 1983. *Regional Scale (1000km) Model of Photochemical Air Pollution, Part 1, Theoretical Formulation*. EPA/600/3-83-035. U.S. Environmental Protection Agency, Environmental Sciences Research Laboratory, Research Triangle Park, NC 27711.

Lamb, R.G., 1984. *Regional Scale (1000km) Model of Photochemical Air Pollution, Part 2, Input Processor Network Design*. EPA/600/3-84-085. U.S. Environmental Protection Agency, Environmental Sciences Research Laboratory, Research Triangle Park, NC 27711.

Law, P.L., Lioy, P.J., Zelenka, M.P., Huber, A.H., and McCurdy, T.R., 1997. Evaluation of a probabilistic exposure model applied to carbon monoxide (pNEM/CO) using Denver personal exposure monitoring data. *Journal of Air & Waste Management Association.* (47):491-500.

Lawson, R., and Snyder, W.H., 1983. *Determination of Good-Engineering-Practice Stack Height: a Fluid Model Demonstration Study for a Power Plant*, EPA/600/3-83-024. U.S. Environmental Protection Agency, Research Triangle Park, NC 27711. 70 pages.

Lee, I.Y., 1987. Numerical simulations of cross-application transport and diffusion. *Boundary-Layer Meteorology*, (39):53-66.

Lee, R.F., Perry, S.G., Cimorelli, A.J., Paine, R.J., Venkatram, A., Weil, J.C., and Wilson, R.B., 1996. AERMOD: The developmental evaluation. (Gryning and Schiermeier, Eds.) *Air Pollution and Its Application XI*, Plenum Press, NY, pages 623-630.

Leighton, P.A., 1961. *Photochemistry of Air Pollution*. Academic Press, New York and London, 279 pages.

Levy, A., 1971. Normal atmosphere: large radical and formaldehyde concentrations predicted. *Science*, 173:141-143.

Levy A., 1972. Photochemistry of the lower troposphere. *Planet, Space Science*, 20:919-935.

Lin, C.J., and Pehkonen, S.O., 1999. The chemistry of atmospheric mercury: A review. *Atmospheric Environment*. 33(13):2067-2079.

Linker, L.C., Stigall, G.E., Chang, C.H., and Donigian, A.S., 1996. Aquatic Accounting : Chesapeake Bay Watershed Model Quantifies Nutrient Loads. *Water Environment and Technology*. 8(1):48-52.

List, R.J., 1954. On the transport of atomic debris in the atmosphere. *Bulletin of American Meteorological Society*. Vol. 35(7):315-325.

List, R.J., 1971. *Smithsonian Meteorological Tables*. Volume 114, Smithsonian Institution Press, City of Washington DC.

Lucas, D.H., 1958. The atmospheric pollution of cities. *International Journal of Air and Water Pollution.* (1):71-86.

Lurmann, F.W., 1991. *Implementation of the 1990 SAPRAC Chemical Mechanism in the Urban Airshed Model*. Prepared for the South Coast Air Quality Management District by Sonoma Technology, Inc., Santa Rosa, CA. 169 pages.

Lurnamm, F.W., Carter, W.P.L., and Coyner, L.A., 1987. *A Surrogate Species Chemical Reaction Mechanism for Urban-Scale Air Quality Simulation Models.* EPA-600/3-87-014, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

Machta, L., and Harris, D.L., 1955. Effects of atomic explosions on weather. *Science*. Vol. 131(3134):75-81.

Martin, D.O., 1971. An urban diffusion model for estimating long term average values of air quality. *J. of the Air Pollution Control Association*. Vol 21(1):16-19.

McCurdy, T., 1994. *Estimating Ozone Exposures in the Chicago Urban Area Using a Second-Generation Probabilistic Version of NEM*. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC.

McElroy, J.L. and Pooler, F., 1968. *St. Louis Dispersion Study Volume II-Analysis.* National Air Pollution Control Administration. Publication Number AP-53. U.S. Department of Health Education and Welfare, Arlington, VA. 51 pages.

Meade, P.J., and Pasquill, F., 1958. A study of the average distribution of pollution around Stayhorpe. *International Journal of Air Pollution.* Pergamon Press. Vol. 1, pages 60-70.

MOI, Work Group II 1982. *Atmospheric Sciences and Analysis, Regional Modeling Subgroup Report*. Report No. 2F-M, Oct. 15.

Moran, M.D., and Pielke, R.A., 1996. Delayed shear enhancement in mesoscale atmospheric dispersion. *8th Joint Conference on the Applications of Air Pollution Meteorology with A&WMA*, American Meteorological Society, Boston, MA, pages 96-103.

National Center for Atmospheric Research, 1983a. *Regional Acid Deposition: Design and Management Plan for a Comprehensive Modeling System*. NCAR/TN-215+PPR, National Center for Atmospheric Research, Boulder, CO, 33 pages.

National Center for Atmospheric Research, 1983b. *Regional Acid Deposition: Models and Physical Process*, NCAR/TN-214+STR, National Center for Atmospheric Research, Boulder, CO, 386 pages.

National Research Council 1983. *Risk Assessment in the Federal Government, Managing the Process*. Washington, D.C., National Academy Press, 191 pages.

National Research Council 1991a. *Rethinking the Ozone Problem in Urban and Regional Air Pollution*. Washington, D.C., National Academy Press, 489 pages.

National Research Council 1991b. *Human Exposure Assessment for Airborne Pollutants: Advances and Opportunities*. Washington, D.C., National Academy Press, 344 pages.

National Research Council 1994. *Science and Judgment in Risk Assessment*. Washington DC, National Academy Press, 651 pages.

Netterville, D.D.J., 1990. Plume rise, entrainment and dispersion in turbulent winds. *Atmospheric Environment*. 24A:1061-1081.

Nohebel, G., 1965. Chimney emissions and immissions: standardization of formulae for estimation of plume rise and ground level concentrations. *International Journal of Air and Water Pollution*. Vol. 9(11):763-766.

Noilhan, J., and Planton, S., 1989. A simple *parameterization of land surface processes for meteorological models. Monthly* Weather Review. 117(3):536-549.

Nordo, J., Eliassen, A., and Saltbones, J., 1974. Large-scale transport of air pollutants. *Advances in Geophysics*. Vol. 18B, Academic Press, pages 137-150.

Novak, J.H., and Reagan, J.A., 1986. A comparison of natural and man-made hydrocarbon emission inventories necessary for regional acid deposition and oxidant modeling. Paper No. 86-30. In: *Proceedings of the 70th Annual APCA Meeting*, Air Pollution Control Association, Pittsburgh, PA, June 1986.

Ottar, B., 1978. An assessment of the OECD study on long range transport of air pollutants (LRTAP). *Atmospheric Environment*. Vol. 12:445-454.

Ott, W., Thomas, J., Mage, D., and Wallace, L. 1988. Validation of the Simulation of Human Activity and Pollutant Exposures (SHAPE) model using paired days from the Denver, CP, carbon monoxide field study. *Atmospheric Environment*. (22):249-287.

Ott, W., Wallace, L., and Mage, D., 2000. Predicting particulate (PM10) personal exposure distributions using a random component superposition statistical model. *Journal of Air & Waste Management Association*. 50:1390-1406.

Owen, B., Edmunds, H.A., Carruthers, D.J., and Singles, R.J., 2000. Prediction of total oxides of nitrogen and nitrogen dioxide concentrations in a large urban area using a new generation urban scale dispersion model with integral chemistry model. *Atmospheric Environment*. 14:397-406.

Pasquill, F., 1961. The estimation of the dispersion of windborne material. *Meteorology Magazine*. Vol. 90(1063):33-49.

Perhac, R.M., 1978. Sulfate regional experiment in northeastern United States: the SURE program. *Atmospheric Environment*. Vol. 12:641-647.

Perry, S.G., 1992a. CTDMPLUS: a dispersion model for sources near complex topography. Part I: technical formulations. *Journal of Applied Meteorology*. American Meteorological Society, Boston, MA, 31(7):633-645.

Perry, S.G., 1992b. CTDMPLUS: a dispersion model for sources near complex topography. Part II: performance characteristics. *Journal of Applied Meteorology*. American Meteorological Society, Boston, MA, 31(7):646-660.

Perry, S.G., Cimorella, A.J., Lee, R.F., Paine, R.J., Venkatram, A., and Weil, J.C., 1994. AERMOD: a dispersion model for industrial source applications. *Proceedings of the 87th Annual Meeting of the AW&MA*, 94-TA23.04, AW&MA, Pittsburgh, PA.

Peters, L.K., Berkowitz, C.M., Garmichail, G.R., Easter, R.C., Fairweather, G., Ghan, S.J., Hales, J.M., Leung, L.R., Pennell, W.R., Potra, R.A., Saylor, R.D., and Tsang, T.T., 1995. The current state and future direction of Eulerian models in simulating the tropospheric chemistry and transport of trace species: a review. *Atmospheric Environmen*t. Vol. 29(2):189-222.

Petersen, W.B., 1982. *Estimating Concentration Downwind from an Instantaneous Puff Release*. EPA 600/3-82-078. Environmental Science Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711. 73 pages.

Petersen, W.B., and Lavdas, L.G., 1986. *INPUFF 2.0 - A Multiple Source Gaussian Puff Dispersion Algorithm User's Guide*. EPA 600/8-86-024. Atmospheric Sciences Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711. 104 pages.

Peterson, E., and Tingey, D., 1980. An estimate of the possible contribution of biogenic sources to airborne hydrocarbon concentrations. *Atmospheric Environment*, 14(1):79-81.

Pierce, T.E, Lamb, B., and Van Meter, A., 1990. Development of a biogenic emissions inventory system for regional scale air pollution models. Paper No. 90-94.3. In: Proceedings of the *83rd Annual AWMA (formerly APCA) Meeting*, June 24-29, 1990. Air and Waste Management Association, Pittsburgh, PA. 16 pages.

Pierson, W.R., Gertler, A.W., Robinson, N.R., Sagebiel, J.C., Zielinske, B., Bishop, G.A., Siedman, D.H., Zweidinger, R.B., and Ray, W.D., 1996. Real-world automotive emissions - summary of studies in the Fort McHenry and Tuscarora Mountain tunnels. *Atmospheric Environment*. 30:2233-2256.

Pitchford, M.L., and Malm, W.C., 1994. Development and application of a standard visual index. *Atmospheric Environment*. 20:1049-1054.

Placet, M., Mann, C.O., Gilbert, R.O., and Niefer, M.J., 2000. emissions of ozone precursors from stationary sources: a critical review. *Atmospheric Environment*. 34:2183-2204.

Pleim, J.E., and Chang, J.S., 1992. A non-local closure model for vertical mixing in the convective boundary layer. *Atmospheric Environment*. 26A:965-981.

Pleim, J.E., and Xiu, A., 1993. Development and testing of a land-surface and PBL model with explicit soil moisture parameterization. Preprints, *Conference on Hydroclimatology*, January 17-22, 1993, Anaheim, CA, American Meteorological Society, Boston, pages 45-51.

Pleim, J.E., and Xiu, A., 1995. Development and testing of a surface flux and planetary boundary layer model for application in mesoscale models. *Journal of Applied Meteorology*. 34:19-32.

Pleim, J.E., Xiu, A., Finkelstein, P.L., and Clarke, J.F., 1997. Evaluation of a coupled land-surface and dry deposition model through comparison to field measurements of surface heat, moisture and ozone fluxes. Preprints, *12th Symposium and Boundary Layers and Turbulence*, July 28-August 1, 1997, Vancouver, British Columbia, Canada. American Meteorological Society, Boston, MA, pages 4787-479.

Pleim, J.E., Finkelstein, P.L., Clarke, J.R., and Ellestard, T.G., 1999. A technique for estimating dry deposition velocities based on similarity with latent hear flux. *Atmospheric Environment*. 33:2257-2268.

Pleim, J.E., Venkatram, A., and Yarmartino, R., 1984. ADOM/TADAR model development program. *Volume 4, Dry Deposition* ERT Document No. P-8980-520, Concord, MA.

Pleim, J.E., Clarke, J.F., Finkelsteim, P.L., Cooter, E.J., Ellestad, T.G., Xiu, A., and Angevine, W.M., 1996. Comparison of measured and modeled surface fluxes of heat, moisture and chemical dry deposition. In *Air Pollution Modeling and Its Application XXI*. Volume 21. S.-E. Gryning and F.A. Schiermeier (Eds.). Plenum Press, New York, NY, pages 613-621.

Poole-Kober, E.M., and Viebrock, H.J., 1993. Fiscal Year 1992 Summary Report of NOAA Atmospheric Sciences Modeling Division Support to the U.S. Environmental Protection Agency. *NOAA Technical Memorandum ERL ARL-203*, Air Resources Laboratory, Silver Spring, MD, June 1993, 85 pages.

Poole-Kober, E.M., and Viebrock, H.J., 1994. Fiscal Year 1993 Summary Report of NOAA Atmospheric Sciences Modeling Division Support to the U.S. Environmental Protection Agency. *NOAA Technical Memorandum ERL ARL-206*, Air Resources Laboratory, Silver Spring, MD, June 1994, 103 pages.

Pooler, F., 1961. A prediction model of mean urban pollution for use with standard wind roses. *International Journal of Air and Water Pollution.* Vol. 4(3/4):199-211.

Possiel, N.C., Milich, L.R., and Goodrich, B.R., (Eds.), 1991. *Regional Ozone Modeling for Northeast Transport (ROMNET) Project Final Report*. EPA-450/4-91-002a. Office of Air Quality Planning and Standards, Research Triangle Park, NC.

Randerson, D., 1979. Review panel on sigma computations. *Bulletin of the American Meteorological Society*. 60:682-683.

Randerson, D. (Editor) 1984. *Atmospheric Science and Power Production.* DE84005177 (DOE/TIC-27601), National Technical Information Service, U.S. Department of Commerce. Springfield, VA 22161. 850 pages.

Rasmussen, R., 1972. What do the hydrocarbons from trees contribute to air pollution? *Journal of the Air Pollution Control Association*, 22(7):537.

Reynolds, S.D., Seinfeld, J.H., and Roth, P.M., 1973. Mathematical modeling of photochemical air pollution -- I. Formulation of the model. *Atmospheric Environment*. (7):1033-1062.

Rodhe, H., 1972. A study of the sulfur budget for the atmosphere over Northern Europe. *Tellus*. Vol. 24:128-138.

Roselle, S.J., and Schere, K.L., 1995. Modeled response of photochemical oxidants to systematic reductions in anthropogenic volatile organic compound and NOx emissions. *Journal of Geophysical Research*. 100(D11):22,929-22,941.

Roselle, S.J., Hanna, A.F., Lu, V., Jang, J-C.C., Schere, K.L., and Pleim, J.E., 1996. Refined photolysis rates for advanced air quality modeling systems., Preprints, *9th Joint Conference on the Applications of Air Pollution Meteorology with A&WMA*, January 28-February 2, 1996, Atlanta, Georgia. American Meteorological Society, Boston, pages 213-216.

Roth, P.M., Reynolds, S.D., Roberts, P.J.W., and Seinfeld, J.H., 1971. *Development of a Simulation Model for Estimating Ground Level Concentrations of Photochemical Pollutants*. Report 71-8AI-21. Systems Applications, Inc., Beverly Hills, CA.

Russell, A., and Dennis, R., 2000. NARSTO critical review of photochemical models and modeling. *Atmospheric Environment*. Vol. 34:2283-2324.

Samson, P.J., Brook, R., and Stillman, S., 1990. *Estimation of Seasonal and Annual Acid Deposition through Aggregation of Three-Day Episodic Periods*. EPA/600/3-90/059, Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC,. 146 pages.

Saxena, P., Hudischewskuyj, A.G., Seigneur, C., and Seinfeld, J.H., 1986. A comparative study of equilibrium approaches to the chemical characterization of secondary aerosols. *Atmospheric Environment*. 20:1471-1483.

Schere, K.L., and Shreffler, J.H., 1983. *Final Evaluation of Urban-scale Photochemical Air Quality Simulation Models*. EPA-600/3-82-094. Environmental Sciences Research Laboratory, Research Triangle Park, NC 27711. 259 pages.

Schere, K.L., and Wayland, E., 1989. *EPA Regional Oxidant Model (ROM 2.0), Evaluation on 1980 NEROS Data Bases*. EPA-600/S3-89/057. U.S. Environmental Protection Agency, Research Triangle Park, NC 27711. 351 pages.

Schiermeier, F.A., Wilson, W.E., Pooler, F., Ching J.K.S., and Clarke, J.F., 1979. Sulfur transport and transformation in the environment (STATE): a major EPA research program. *Bulletin of American Meteorological Society*. Vol. 60(11):1303-1312.

Schock, M.R., and Weber, S.F., 1984. *MSPUFF Mesoscale Air Quality Computer Modeling System*. Division of Environmental Waste Management and Research. North Dakota State Department of Health. Bismarck, ND.

Schrenk, H.H., Heimann, H., Clayton, G.D., Gafafer, W.M., and Wexler, H., 1949. *Air Pollution in Donora, PA*. Public Health Bulletin No. 306. U.S. Public Health Service, Washington DC, 173 pages.

Schuck, E.A., Altshuller, A.P., Barth, D.S., and Morgan, G.B., 1970. Relationship of hydrocarbons to oxidants in ambient atmospheres. *Journal of Air Pollution Control Association*. Volume 20, No. 5:297-302.

Scire, J.S., Yamartino, R.J., and Fernau, M.E., 2000a. *A User's Guide for the CALMET Meteorological Model*. Earth Tech, Concord, MA, http://www.src.com (accessed March 2000).

Scire. J.S., Strimaitis, and Yamartino, R.J., 2000b. *A User's Guide for the CALPUFF Dispersion Model*. Earth Tech, Concord, MA, http://www.src.com (accessed March 2000).

Scire, J.S., Lurmann, F.W., Bass, A., and Hanna, S.R., 1984. *Users Guide to MESOPUFF II Model and Related Processor Programs*. Environmental Research and Technology, Inc.,, Concord, MA, Prepared for Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Scorer, R.S., 1959. The behavior of chimney plumes. *International Journal of Air Pollution*. (1):198-220.

Seibert, P., Beyrich, F., Gryning, S-E., Joffre, S., Rasmussen, A., Tercier, P., 2000. Review and intercomparison of operational methods for the determination of the mixing height. *Atmospheric Environment*. Vol. 34:1001-1027.

Seigneur, C., 1982. A model of sulfate aerosol dynamics in atmospheric plumes. *Atmospheric Environment*. 16:2207-2228.

Seigneur, C., Praised, P., Hope, PK, Grosjean, D., 1999. Modeling atmospheric particulate matter. *Environmental Science & Technology*. Vol. 3:80A-86A.

Seinfeld, J.H., 1988. Ozone air quality models: a critical review. *Journal of the Air Pollution Control Association*. 14:616-645.

Shankar, W.H., and Binkowski, F.S., 1994. Sulfate aerosol wet deposition in a three-dimensional Eulerian air quality modeling framework. Abstracts, *4th International Aerosol Conference*, August 29-September 2, 1994, Los Angeles, CA, Richard C. Flagan (Ed.), American Association for Aerosol Research, Cincinnati, OH, page 417.

Shreffler, J.H., 1978. Detection of centripetal heat-island circulations from tower data in St. Louis. *Boundary-Layer Meteorology*, (15):229-242.

Skalew, R.C., Fabrick, A.J., and Prager, J.E., 1971. *A Particle-In-Cell method for Numerical Solution of the Atmospheric Diffusion Equation and Application to Air Pollution Problems*. Final Report (NTIS PB-299-290). Systems, Science and Software, Inc., La Jolla, CA.

Smith, M.E., 1984. Review of the attributes and performance of 10 rural diffusion models. *Bulletin of the American Meteorological Society*. (65):554-558.

Snyder, W.H., 1972. Similarity criteria for the application of fluid models to the study of air pollution meteorology. *Boundary-Layer Meteorology*. (3):113-134.

Snyder, W.H. and Lawson, R.E., jr., 1976. Determination of a necessary height for a stack close to a building - a wind tunnel study. *Atmospheric Environment*. (10):683-691

Snyder, W.H., Thompson, R.S., Eskridge, R.E., Lawson, R.R., Castro, I.P., Lee, J.T., Hunt, J.C.R, Ogawa, Y., 1985. The structure of strongly stratified flow over hills: dividing-streamline concept. *Journal of Fluid Mech*., (152):249-288.

Stauffer, D.R., and Seaman, N.L., 1990. Use of four-dimensional data assimilation in a limited-area mesoscale model. Part I: Experiments with synoptic-scale data. *Monthly Weather Review*, Vol. 118:1250-1277.

Stauffer, D.R., Seaman, N.L., and Binkowski, F.S., 1991. Use of four-dimensional data assimilation in a limited-area mesoscale model. Part II: Effects of data assimilation within the planetary boundary layer. *Monthly Weather Review*, Vol. 119:734-754.

Stern, A.C., 1976, 1977. *Air Pollution. Volume 1. Air Pollutants, Their Transformation and Transport; Volume 2. The Effect of Air Pollution; Volume 3. Measuring, Monitoring, and Surveillance of Air Pollution; Volume 4. Engineering Control of Air Pollution; Volume 5. Air Quality Management*. Academic Press, New York.

Strothmann, J.A., and Schiermeier, F.A., 1979. *Documentation of the Regional Air Pollution Study (RAPS) and Related Investigations in the St. Louis Air Quality Control Region.* EPA-600/4-79-076. Environmental Science Research Laboratory, Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, NC. 715 pages.

Suck, S.H., and Brock, J.R., 1979. Evolution of atmospheric aerosol particle size distributions via Brownian coagulation: Numerical simulation. *Journal of Aerosol Science*. 10:581-590.

Sutton, O.G., 1947. The theoretical distribution of airborne pollution from factory chimneys. *Quarterly Journal of the Royal Meteorology Society*. (73):426-436.

Taylor, G.I., 1945. Dynamics of a Mass of Hot Gas Rising in the Air, *USAEC Report MDDC-919 (LADC-276)*, Los Alamos Scientific Laboratory.

Trainer, M., Williams, E., Parrish, E., Buhr, M., Allwind, E., Westberg, H., Fahsenfeld, R., and Liu, S., 1987. Models and observations of the impact of natural hydrocarbons on rural ozone. Nature, 329:705.

Tsang, T.H., and Brock, J.R., 1982. Aerosol coagulation in the plume from a cross-wind line source. Atmospheric Environment. 16:2229-2235.

Tsang, T.H., and Brock, J.R., 1983. Simulation of condensation aerosol growth by condensation and evaporation. Aerosol Science and Technology. 2:311-320.

Turner, D.B., 1961. Relationships between 24-hour mean air quality measurements and meteorological factors in Nashville, Tennessee. *Journal of Air Pollution Control Association.* (11):483-489.

Turner, D.B., 1964. A diffusion model for an urban area. *Journal of Applied Meteorology*. Vol. 3(2):83-91.

Turner, D.B., 1967. *Workbook of Atmospheric Dispersion Estimates*. Public Health Service Publication No. 999-AP-26. U.S. Public Health Service, Cincinnati, OH. 84 pages.

Turner, D.B., 1979. Atmospheric dispersion modeling. a critical review. *Journal of Air Pollution Control Association*. Volume 29(5):502-519.

Turner, D.B. and Edmisten, N.G., 1968. *St. Louis SO2 Dispersion Model Study - Description of Basic Data*. (Unpublished report, Division of Meteorology, NAPCA). Atmospheric Sciences Modeling Division, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

Turner, D.B. and Irwin, J.S., 1983. Comparison of sulfur dioxide estimates from the model RAM with St. Louis measurements. *Air Pollution Modeling and Its Application II*, (Edited by C. De Wispelaere), Plenum Press, pages 695-707.

Turner, D.B., and Irwin, J.S., 1985. The relation of urban model performance to stability. *Air Pollution Modeling and Its Application IV*, (Edited by C. De Wispelaere), Plenum Press, pages 721-732.

Turner, D.B. and Novak, J.H., 1978. *User's Guide for RAM, Volume 1, Algorithm Description and Use*. EPA-600/8-78-016a. Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, 70 pages.

Turner, D.B., Chico, T., and Catalano, J.A., 1986. *TUPOS - A Multiple Source Gaussian Dispersion Algorithm Using On-Site Turbulence Data*. EPA-600/8-86-010 [NTIS PB 86-181 310/AS]. U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, 171 pages.

Turner, D.B., Irwin, J.S., and Busse, A.D., 1985. Comparison of RAM model estimates with 1976 St. Louis RAPS measurements of sulfur dioxide. *Atmospheric Environment*. (19):247-253.

Turner, D.B., Zimmerman, J.R., and Busse, A.D., 1971. An evaluation of some climatological models. Paper presented at Third Meeting of the NATO/CCMS Panel on Modeling. Paper published in Appendix E of *User's Guide for the Climatological Dispersion Model*. EPA-R4-73-024. Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, pages 107-131.

Turner, D.B., Bender, L.W., Pierce, T.E., and Petersen, W.B., 1989. Air quality simulation models from EPA. *Environmental Software*. Vol. 4(2):52-61.

U.S. Environmental Protection Agency 1971. *Air Quality Criteria for Nitrogen Oxides*. AP-84. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC 27711.

U.S. Environmental Protection Agency, 1976. *Formation and Transport of Oxidants Along Gulf Coast and in Northern U.S.* EPA-450/3-76-033. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC 27711, 481 pages.

U.S. Environmental Protection Agency 1977. *User's Manual for Single-Source (CRSTER) Model*. EPA-450/2-77-013, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards. Research Triangle Park, NC 27711, 303 pages.

U.S. Environmental Protection Agency 1978a. *User's Guide for RAM*. EPA-600/8-78-016a, Environmental Sciences Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

U.S. Environmental Protection Agency 1978b, 1986, 1987, 1995. *Guideline on Air Quality Models*. EPA-450/2-78-027, [This Guideline has been updated several time: 1986 as EPA-450/2-78-027R; 1987 as EPA/450/2-78/027R-SUPPL-A; 1993 as EPA-450/2-78-027R-B, and 1995 it was updated and incorporated into Appendix W to 40 CFR Part 51], U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards. Research Triangle Park, NC 27711.

U.S. Environmental Protection Agency 1979. *Industrial Source Complex (ISC) Dispersion Model User's Guide*. Volume I EPA-450/4-79-030 and Volume II EPA-450/4-79-031, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

U.S. Environmental Protection Agency 1980. *Guideline for Applying the Airshed Model to Urban Areas*. EPA-450/4-80-020, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards Research, Triangle Park, NC 27711.

U.S. Environmental Protection Agency 1985a. *Fluid Modeling Demonstration of Good-Engineering-Practice Stack Height in Complex Terrain*. EPA/600/3-85-022, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, 89 pages.

U.S. Environmental Protection Agency, 1985b.*Compilation of Air Pollutant Emission Factors, Vol. 1: Stationary Point and Area Sources*, AP-42, 4th Edition, GPO No. 055-000-00251-7, U.S. Environmental Protection Agency, Research Triangle Park, NC.

U.S. Environmental Protection Agency 1988a. *User's Guide for Executing OZIPM-4 (Ozone Isopleth Plotting with Optional Mechanisms, Version 4) with CMB-IV (Carbon-bond Mechanisms-IV) or Optional Mechanisms*. Volume I, description of the ozone isopleth plotting package, version 4. EPA/600/8-88-073a, Atmospheric Sciences Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, 240 pages.

U.S. Environmental Protection Agency, 1988b. *Development and Testing of the CBM-IV (Carbon-Bond Mechanism) for Urban and Regional Modeling*. EPA/600/3-88/012, Atmospheric Sciences Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, 431 pages.

U.S. Environmental Protection Agency, 1992. *Guidelines for Exposure Assessment*. EPA/600Z-92/001. Federal Register 57: 22888 - 22938, 169 pages.

U.S. Environmental Protection Agency 1995. *Users Guide for the Industrial Source Complex (ISC3) Dispersion Models, Volume I - User Instructions.* EPA-454/B-95-003a. Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, 320 pages.

U.S. Environmental Protection Agency 1996. *EPA At Research Triangle Park. Twenty Five Years of Environmental Protection*. Office of Administration and Resources Management, Research Triangle Park, NC 27711, 50 pages.

U.S. Environmental Protection Agency 1997. *Mercury Study Report to Congress. Volume I: Executive Summary*. EPA-452/R-97-003. Office of Air Quality Planning and Standards and Office of Research and Development, Research Triangle Park, NC.

U.S. Environmental Protection Agency 1998a. *Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units - Final Report to Congress.* Volume 1. EPA-453/R-98-004a. Office of Air Quality Planning and Standards, Research Triangle Park, NC.

U.S. Environmental Protection Agency, 1998b. *Guidelines for Ecological Risk Assessment*, Federal Register 63(93): 26846-26924, 188 pages.

U.S. Weather Bureau 1955. *Meteorology and Atomic Energy*. AECU 3066, U.S. Atomic Energy Commission, Washington DC, 169 pages.

Valente, R., J., and Thornton, F.C., 1993. Emissions of NO from soil at a rural site in central Tennessee. *Journal of Geophysical Research*. 98:16745-16753.

Warren, D.R., and Seinfeld, J.R., 1985. Simulation of aerosol size distribution evolution in systems with simultaneous nucleation, condensation, and coagulation. *Aerosol Science and Technology*. 4:31-43.

Wayne, G., Kokin, A., and Wiesburd, M.I., 1973. *Controlled Evaluation of the Reactive Environmental Simulation Model (REM)*, EPA-R4-73-013, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Weil, J.C., 1988. Plume Rise. In: *Lectures on Air Pollution Modeling* (A. Venkatram and J.C. Wyngaard (Eds.). American Meteorological Society, Boston, MA, pages 119-166.

Weil, J.C., 1992. Updating the ISC model through AERMIC. *Proceedings of the 85th Annual Meeting of the AW&MA*, 92-100.11, AW&MA, Pittsburgh, PA.

Weil, J.C., and Brower, R.P., 1984. An updated Gaussian plume model for tall stacks. *Journal of Air Pollution Control and Association*. (34):818-827.

Weil, J.C., and Corio, L.A., 1988. *A Modification of the PPSP Dispersion Model for Highly Buoyant Plumes*. PPRP-MP-60, Maryland Power Plant Research Program, 46 pages.

Weisburd, M., Wayne, G., Danchick, R., Kokin, A., and Stein, A., 1971. *Development of a Simulation Model for Estimating Ground Level Concentrations of Photochemical Pollutants*. Final Report. TM-(L), 4673/000/00. System Development Corp., Santa Monica, CA.

Wesely, M.L., Cook, D.R., and Hart, R.L., 1985. Measurement and parameterization of particulate sulfur dry deposition over grass. *Journal of Geophysical Research*. 90(D1):2131-2143.

Whitby, K.T., 1978. The physical characteristics of sulfur aerosols. *Atmospheric Environment*. 12:135-159.

Whitby, K.T., 1981. Determination of aerosol growth rates in the atmosphere using lumped aerosol dynamics. *Journal of Aerosol Science*. 12:174-178.

Whitby, E.R., 1985. *The Modal Aerosol Dynamics Model. Part 1. Solution of the Internal Terms of the General Dynamic Equation of Aerosols*. Report to the U.S. Environmental Protection Agency. Department of Mechanical Engineering. University of Minnesota, Minneapolis, MN.

Whitby, E.R., 1990. *Modal Aerosol Dynamics Modeling*. Ph.D. thesis, University of Minnesota, Minneapolis, MN.

Whitby, E.R., McMurray, P.H., Shankar, U., and Binkowski, F.S., 1991. *Modal Aerosol Dynamics Modeling*. EPA/600/3-91-030. [NTIS PB 91-161-729]. Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC 27711.

Whitten, G.Z., Hogo, H., and Killus, J.P., 1980. The carbon-bond mechanism: a condensed kinetic mechanism for photochemical smog. *Environmental Science & Technology*. 14:690-700.

Wilkins, E.T., 1953. *Air Pollution and the London Fog of December 1952.* The Royal Sanitary Institute. Vol. 74(1):1-15.

Wilkinson, J.G., Loomis, G.F., McNally, D.E., Emigh, R.A., and Tesche, T.W., 1994. *Technical Formulation Document: SARMAP/LMOS Emissions Modeling System (EMS-95)*, AG90/TS26, prepared by Alpine Geophysics, Pittsburgh, PA. For the Lake Michigan Air Directors Consortium and California Air Resources Board.

Williams, E.J., Guenther, A., Fehsenfeld, F.C., 1992. An inventory of nitrate oxide emissions from soils in the United States. *Journal of Geophysical Research.* 97(D7):7511-7519.

Willis, G.E., and Deardorff, J.W., 1976. A laboratory model of diffusion in to the convective planetary boundary layer. *Quarterly Journal of the Royal Meteorological Society.* (102):427-445.

Willis, G.E., and Deardorff, J.W., 1978. A laboratory study of dispersion from an elevated source within a modeled convective planetary boundary layer. *Atmospheric Environment.* 12(6-7):1305-1312.

Willis, G.E., and Deardorff, J.W., 1981. A laboratory study of dispersion from a source in the middle of the convectively mixed layer. *Atmospheric Environment.* 15(2):109-117.

Wilson, W.E., 1978. Sulfates in the atmosphere: a progress report on project MISTT. *Atmospheric Environment*. Vol. 12:537-547.

Young, J.O., Alssa, M., Boehm, T.L., Coats, C.J., Elchinger, J.R., Roselle, S.J., Van Meter, A.R., Wayland, R.A., and Pierce, T.E., 1989. *Development of the Regional Oxidant Model, Version 2.1*. EPA/600/3-89/044. (NTIS PB89-194252). EPA, Office of Research and Development, Atmospheric Research and Exposure Assessment laboratory, Research Triangle Park, NC.