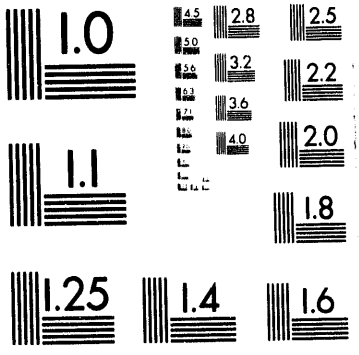
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Global Tropospheric Chemistry Models for Radiatively Important Trace Species: Design and Research Recommendations

W. R. Barchet
A. J. Brothers
C. M. Berkowitz
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S. J. Ghan
R. D. Saylor

December 1993

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GLOBAL TROPOSPHERIC CHEMISTRY MODELS FOR RADIATIVELY IMPORTANT TRACE SPECIES: DESIGN AND RESEARCH RECOMMENDATIONS

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MASTER

EXECUTIVE SUMMARY

This report summarizes the recommendations of a panel of atmospheric scientists, mathematicians, and computer scientists regarding design considerations for a global tropospheric chemistry modeling system (GTCMS). These recommendations are a preliminary step toward developing a model that addresses radiatively important trace species. Many global tropospheric chemistry models are being currently developed by various entities in academia, institutes in Europe, federal laboratories, and private industry, and one or more of these may serve as the initial basis for the modeling system as conceived in this report.

The GTCMS described in this report is based on the current state of atmospheric chemistry modeling science. The GTCMS design requires the use of the most advanced parameterizations consistent with complete mathematical descriptions of the physical and chemical processes acting on the radiatively important trace species. Consisting of a computational framework and chemical transport models for three classes of radiatively important trace species, the GTCMS will provide global simulations of the distributions of important tropospheric trace gases and aerosols. This report focuses on the atmospheric modeling aspects of the design of a GTCMS and does not address the computer engineering aspects of implementing the design.

Three chemical transport models in the GTCMS, designed to address the modeling needs of short-lived (O_3, NO_x, CO, nonmethane hydrocarbon compounds), long-lived (CH_4), and aerosol species that are radiatively important and reactive in the troposphere, are presented. These three models are needed to balance the process detail essential to simulating the tropospheric distribution of the concentrations of these classes of radiatively important trace species. Processes that directly affect their concentrations are treated in considerable detail, whereas processes that are of lesser importance use highly parameterized representations.

Central to the development of all three models is the creation of new chemical mechanisms that deal with the range of chemical mixtures expected on the global scale. That is, these mechanisms must handle the high concentrations found in urbanized areas as well as the very low concentrations found in

the remote regions of the globe. Research is needed to determine whether the short-lived species model can provide the global concentration fields of the important gas- and aqueous-phase oxidants needed by the long-lived and aerosol species models. Considerable attention must be given to the upper boundary condition (stratosphere-troposphere exchange), especially for the short-lived and long-lived species models because the stratosphere may be a dynamic source, as well as a sink, for these species.

Global-scale computations with the GTCMS will require significant resources. The framework for the model must accommodate or be based on parallel computation as much as possible. The Models-3 system being developed by the U.S. Environmental Protection Agency could provide that framework. Only through the efficient use of computational resources can the GTCMS achieve the performance needed for policy applications. Efficiencies can be gained through the development of new solvers for the sets of stiff differential equations required by the chemical mechanisms, advanced algorithms for using vector and parallel coding, and new parameterizations for the less-critical process modules. Advanced methods for dealing with the size distributions of aerosols and cloud droplets must be developed and tested before the aerosol model can be implemented. Specialized coding to enable uncertainty and sensitivity analyses must be incorporated as the models are built; autodifferentiation needs to be demonstrated as a useful tool.

By developing the three chemical transport models around the same computational framework, input data on topography and land use, emissions inventories and models, and meteorological models can have the same format and structure. Likewise, output data from the three models will have compatible formats and structures to simplify the processing of GTCMS output by other models.

Decision analysis methodologies were applied to the design of the three chemical transport models. Optimal selection of process modules in the model was obtained through a decision analysis method called multiattribute utility theory. This method quantified the decision-making process, forcing participants to carefully examine their choices. A decision-support tool, based on multiattribute utility theory and value-based planning, is presented.

Application of this tool at two workshops led to the definition of research activities needed to support the development and implementation of the GTCMS.

This research was funded through interagency agreement DW89935712-01-0 between the U.S. Environmental Protection Agency and the U.S. Department of Energy. The technical team responsible for the workshops to gather and then synthesize the research for the GTCMS presented here is from the Earth and Environmental Sciences Center at Pacific Northwest Laboratory,[a] Richland, Washington.

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• Ken Pickering	NASA	Air quality/mesoscale modeling
• Kenneth Reckhow	Duke	Decision and operational analysis
• Doug Rotman	LLNL	Global atmospheric chemical modeling
• Fred Vukovitch	SAIC	Meteorology, air quality data analysis
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ACRONYMS AND CHEMICAL FORMULAS

AREAL	Atmospheric Research and Exposure Assessment Laboratory
CCN	cloud condensation nuclei
CFC	the family of chlorofluorocarbon compounds
CH_4	methane
CO	carbon monoxide
CO_2	carbon dioxide
CTM	chemical transport model(s)
DMS	dimethylsulfide (CH_3SCH_3)
EC	elemental carbon aerosol
EPA	U.S. Environmental Protection Agency
GCM	general circulation or climate model(s)
GTCMS	global tropospheric chemistry modeling system
GWP	greenhouse warming potential
H_2O_2	hydrogen peroxide
H_2SO_4	sulfuric acid
HC	hydrocarbon compounds
HCHO	formaldehyde
HNO_3	nitric acid
HO_2	hydroperoxyl radical
^{85}Kr	krypton isotope, atomic weight 85
LANL	Los Alamos National Laboratory
LLNL	Lawrence Livermore National Laboratory
MAU	multiattribute utility (theory)
Models-3	EPA's advanced modeling system
MSA	methanesulfonic acid (CH_3SO_3H)
N_2O	nitrous oxide
N_2O_5	dinitrogen pentoxide
NASA	National Aeronautics and Space Administration
NCSC	North Carolina Supercomputing Center
NH_3	ammonia
NH_4NO_3	ammonium nitrate
$(NH_4)_2SO_4$	ammonium sulfate
NMHC	nonmethane hydrocarbon compounds

NO_x	nitrogen oxides, usally NO and nitrogen dioxide (NO_2)
NO_y	various nitrogen oxides, usually NO, NO_2, HNO_3, NO_3 aerosols, and peroxyacetylene nitrate (PAN)
ODE	ordinary differential equation
OH	hydroxyl radical
O_3	ozone
^{210}Pb	lead isotope, atomic weight 210
PNL	Pacific Northwest Laboratory
QSSA	quasi-steady-state approximation
RFP	request for proposal(s)
^{222}Rn	radon isotope, atomic weight 222
RITS	radiatively important trace species
S(IV)	sulfur, valence state IV
S(VI)	sulfur, valence state VI
SAIC	Science Applications International Corporation
SO_2	sulfur dioxide
U-IA	University of Iowa
U-KY	University of Kentucky
U-NC	University of North Carolina
U-NELA	University of Northeastern Lousiana
USC	University of Southern California
VBP	value-based planning

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1.0 INTRODUCTION

Tropospheric chemistry plays multiple roles in the global climate system. Some chemically reactive tropospheric species are radiatively active, either as greenhouse gases (CH_4, O_3) or as light-scattering aerosols (sulfate, nitrate, and elemental carbon aerosols). Although the concentrations of these reactive greenhouse gases are much lower than those of the nonreactive species (CO_2 and water vapor), they are much more effective per molecule. Together, the reactive greenhouse gases contribute some 20% of the total greenhouse warming potential (GWP) (IPCC 1990). Kaufmann et al. (1991) have estimated that, even though coal and oil combustion emit 120 times as many CO_2 molecules as SO_2 molecules and even though sulfate aerosols have atmospheric lifetimes several orders of magnitude shorter than CO_2, the cooling effect of sulfate aerosols scattering sunlight both directly and indirectly as cloud condensation nuclei (CCN) is comparable to the warming effect of the CO_2 emitted in the same combustion process.

Other tropospheric chemical species influence the planetary radiation balance by acting as aerosol precursors (SO_2, NH_3, dimethylsulfide [DMS], methanesulfonic acid [MSA], NO_x, nonmethane hydrocarbons [NMHC]) or by altering the chemical balance that determines the concentration of other greenhouse gases (e.g., CO, NO_x, NMHC influence CH_4, O_3). Most CCN are composed of sulfates arising from the oxidation of DMS and SO_2 (Twomey 1971), and most direct scattering by aerosols is attributed to sulfate (Charlson et al. 1992). The concentration of the hydroxyl radical (OH), which controls the lifetime and, hence, influences the concentration of the reactive greenhouse gases and aerosol precursors, is itself influenced by the concentrations of CO, NO_x, CH_4, and NMHC (Wayne 1991). The GWP of NO_x constitutes 6% of the total GWP over a 100-year period, solely through its role in altering the concentration of tropospheric O_3 (IPCC 1990). The GWP of both CO and NMHC is less than 1%, but CO and NMHC may alter the planetary radiation balance by influencing OH and, hence, the oxidation of SO_2 to form sulfate aerosols.

Changes in the Earth's climate could significantly affect regional and global concentrations of trace species that are criteria pollutants regulated by the U.S. Environmental Protection Agency (EPA). The policy community also

needs to know how changes in global natural and anthropogenic emissions of greenhouse gases, particulate aerosols, and aerosol precursors will affect the distribution and concentrations of these pollutants. This report maps out one path for obtaining this information.

1.1 OVERVIEW OF THE GLOBAL TROPOSPHERIC CHEMISTRY MODELING SYSTEM

Presented in this section is an overview of the design of the global tropospheric chemistry modeling system (GTCMS) for radiatively important trace species (RITS).[a] The term "modeling system" describes a set of independent models that, taken together, simulate a wide range of chemical and physical processes acting on RITS in the troposphere. This system consists of a core chemical transport model (CTM) designed to provide global simulations for short-lived RITS (NO_x, O_3, CO, NMHCs) and to provide global distributions of important tropospheric oxidants (OH, hydroperoxyl radical [HO_2], O_3, and H_2O_2). These oxidant concentration fields are used by the other CTMs within the system; namely, a global model for long-lived RITS and a global model for aerosols. These secondary models are dependent on the core model for detailed descriptions of oxidant fields, so that these models can be run for long-term simulations without exacting the full computational cost of the complex, short-lived species CTM.

The GTCMS will be one component of a larger modeling system for evaluating policy decisions that may change the global concentrations and distributions of RITS. Those policy decisions must account for the potential climatic, social, and economic impacts of those changes and for the changes in the concentrations of criteria pollutants that may occur under other climatic conditions. Other models, not part of the scope of the design of the GTCMS but components of the larger policy-modeling system, are also needed to fully address key policy issues.

Figure 1.1 illustrates the linkages between the GTCMS and the other components of the larger modeling system that will be needed to carry out

---

(a) RITS include chemical species that are radiatively active (e.g., CH_4, N_2O O_3, CO_2, aerosols), precursors of the radiatively active species (e.g., NO_x, NMHC, SO_2), and species that affect the chemical balances of these species (e.g., CO, OH, H_2O_2).

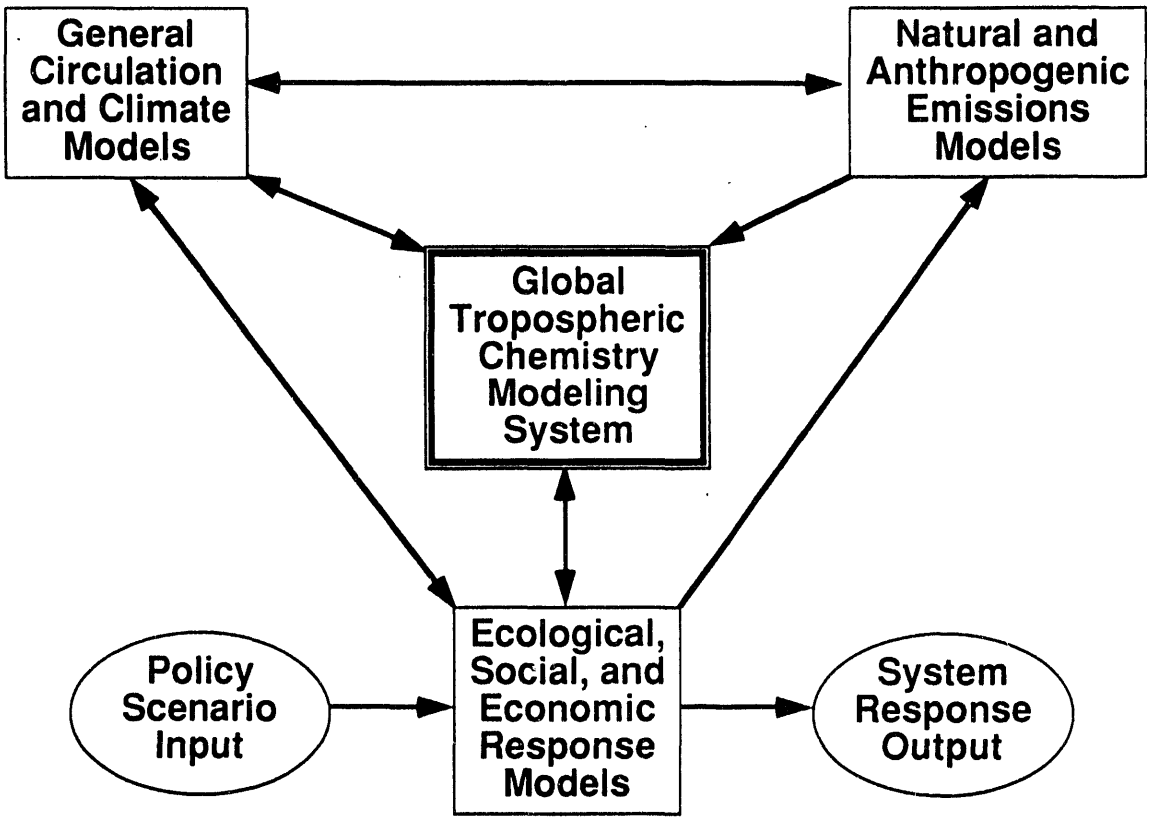

FIGURE 1.1. Relationship of the Global Tropospheric Chemistry Modeling System to the Components of a Global Policy Analysis System

policy analysis. The interconnecting lines indicate the dominant information flows between these models. Policy decisions lead to various scenarios that affect social and economic responses. One or more emissions models respond to the effects of the policy scenario and provide the input of chemical species to the GTCMS. The emissions models predict how policy decisions will affect global emissions of a wide range of compounds that affect RITS concentrations. General circulation or climate models (GCMs) provide global meteorological fields, consistent with current or altered climates, that drive the transport and control the chemistry of RITS and other reactive species in the GTCMS and the emissions of RITS and other reactive species. Ecological, social, and economic response models provide land-use and surface-cover information to both the GTCMS and the GCM and pass this information along with economic and demographic information to the emissions models. The double-arrow interconnecting lines indicate feedback paths that further couple the models.

Ecological response models depend on climate, air quality, and radiation reaching the surface. The GCMs depend on the distribution of radiative forcing caused by the distribution of RITS as provided by the GTCMS. The response of ecological, social, and economic systems to changes in climate and RITS concentrations will result in a system response that policymakers can link to the input policy scenario. The GTCMS must interface as seamlessly as possible with these other models, so that policymakers can address that wide range of policy questions whose answers depend, in part, on changes in RITS concentrations.

The GTCMS described in this report is founded on the current state of atmospheric chemistry modeling science. Whenever possible, the GTCMS design calls for the use of the most advanced parameterizations consistent with complete mathematical descriptions of the physical and chemical processes acting on the RITS. The GTCMS and the larger modeling system of which it is a part will rely heavily on high-performance computing to achieve the timely turnaround needed to support policy analysis and decision making. The computational system will have the memory and computational speed required to handle the complex algorithms in the GTCMS. This report focuses on the atmospheric modeling aspects of the design of the GTCMS and does not address the computer engineering aspects of implementing the design. However, the overall structure and software engineering development of the GTCMS may benefit significantly by taking advantage of the modeling environment as envisioned by the Models-3 development effort (Dennis and Novak 1991).

The conceptual design of the GTCMS as presented in this report does not necessarily require the construction of an entirely new three-dimensional chemistry model. A brief synopsis of current global tropospheric chemistry models is presented in Appendix A. These models include recent efforts at the Geophysical Fluid Dynamics Laboratory (Kasibhatla et al. 1993), Harvard University (Jacob et al. 1993a, 1993b; Balkanski et al. 1993), Max Planck Institute for Chemistry (Dentener and Crutzen 1993; Langner and Rodhe 1991), Lawrence Livermore National Laboratory (Penner et al. 1993), Pacific Northwest Laboratory (PNL) (Easter et al. 1993), and Los Alamos National Laboratory (Tie et al. 1992). Additional work in this area is ongoing at the National Center for Atmospheric Research, Cambridge University, Massachusetts Institute of

Technology, Atmospheric and Environmental Research, Inc., and Goddard Institute for Space Studies. However, because these efforts are in relatively early developmental stages, little or no published information is available. It is conceivable that one or more of these current tropospheric chemistry models may be adequate to serve as the initial basis for the GTCMS as conceived in this report. None of the current global tropospheric chemistry models has exactly the structure or includes all of the processes that are recommended here for the GTCMS. It is not the purpose of this report to recommend a specific model or models on which to base the GTCMS.

1.2 THE ROLE OF DECISION ANALYSIS

The task of designing a GTCMS is extremely complex and must take many considerations into account. Among these are: what level of scientific detail should be included in the model; does the current state of science allow the GTCMS to be developed using existing models; how much risk should be accepted in the development of new modeling tools as part of the GTCMS; is additional research needed for GTCMS development? If additional research is needed, then what are the critical research needs and what components of the GTCMS can be developed with existing science? These considerations illustrate the complexity of the design task and point to the need for a methodology that leads to a logically defensible design of the GTCMS and to the definition of the research and development needed to achieve it.

Decision analysis (see, for example, Keeney and Raiffa 1976 or von Winterfeldt and Edwards 1986) was chosen as the methodology because it makes explicit the rationale by which decisions are reached and promotes clarity in the thought process. Decision analysis is a structured, theoretically grounded process for decision making, fostering a conscientious recognition of the factors that enter into decision making (see von Neumann and Morgenstern 1947 and Savage 1954). Applications of decision analysis in this project were threefold: design of the GTCMS, identification of research needs, and development of a decision-support system for the prioritization of research needs and optimization of research resources. Two decision analysis tools were used at key points in the process: multiattribute utility theory and value-based planning.

Multiattribute utility theory was applied to the selection of module options that form the design of the GTCMS. By setting various design objectives, several versions of each CTM were constructed from a menu of options for each of the major process modules. Evaluation of those versions against specific design criteria resulted in selecting a version for each class of CTM that was considered of greatest value to the GTCMS.

Through a comparison of the current state of development and the design goals for the various modules in the GTCMS, research needs were identified. Based on these needs, research activities were defined that would produce a GTCMS in a 3-year time period. Multiattribute utility theory, combined with value-based planning, yields a decision-support tool for prioritizing those research activities. Illustration of the application of this decision-support tool to the creation of an optimal portfolio of recommended research activities carried the decision process to its logical conclusion. The decision-support tool described herein can be updated and modified as necessary to accommodate new applications. Thus, the methodology described in this report provides a tool that can be used in many settings other than the design of the GTCMS.

1.3 APPROACH

This report is the result of the synthesis and expansion of information gathered at two workshops led by the research team at PNL. Participants at the workshops included technical specialists in various aspects of atmospheric chemistry modeling from universities, scientific consulting firms, and federal laboratories and agencies (see Acknowledgments).

The first workshop focused on defining the attributes and capabilities of the modeling system. Here, the distinction was defined between the GTCMS and the larger modeling system of which it is a part. Lists were developed of various approaches to simulating processes and data needs for input and output from the various components of the GTCMS. After the workshop, the PNL team took these data and developed a decision analysis method to select which options for the various process modules were most suitable for the GTCMS. The first workshop provided a broad overview of what the GTCMS must do but did not

address design details. A summary of this workshop was distributed to the participants.

During preparation of the second workshop, it was recognized that more than one CTM was needed in the GTCMS to deal with the differences between different classes of RITS. Therefore, the second workshop was structured around developing specific designs for each class of CTM using decision analysis. Many of the participants of the first workshop participated in the second, along with other technical specialists. Participants selected those options to the various process models that would yield the best CTM for its class of RITS and specified the research needed to implement those options. Multiattribute utility theory was used to establish priority among the suggested research activities. A summary of this workshop was also provided to the participants.

Following the second workshop, the PNL team synthesized the selected design options into a description of the modular components needed for each class of CTM, expanded the descriptions of research activities that were outlined, and extended the methodology used in the research prioritization to include value-based planning. An illustrative example is presented of the application of value-based planning to the creation of portfolios of recommended research that makes optimal use of available resources.

In this project, the exercises of the first and second workshops illustrated the application of multiattribute utility theory to the design of the GTCMS and to the ranking of research needs for GTCMS development. The process was carried further in this report by describing how value-based planning leads to the optimal construction of research portfolios for different resource levels. These processes are generally applicable to many decision processes. Application of multiattribute utility theory and value-based planning requires input from many perspectives and areas of expertise. Significant attention must be given to setting a clear goal for the process. Definition of evaluation criteria and performance objectives through a group process helps to ensure that the criteria are comprehensive, measurable, and inclusive of all factors that must be considered. More time should be allocated to this process than was available at our workshops.

1.4 STRUCTURE OF THIS REPORT

Section 2.0 presents recommendations for the design of three interrelated versions of the CTMs, along with recommendations of the research activities needed to support the development and implementation of the GTCMS. Section 3.0 presents an overview of the decision process that was applied to the design of the GTCMS and of the applications of decision analysis to assigning priorities to research activities and to defining optimal portfolios of research activities. References follow in Section 4.0.

Appendix A gives a brief overview of current CTMs. Appendix B presents the matrix of options for the various process modules included in the GTCMS. Expanded descriptions of research activities recommended to support the development and implementation of the GTCMS design are given in Appendix C.

2.0 DESIGN AND RESEARCH RECOMMENDATIONS

Recommendations for the design of the GTCMS and for the research needed to support its development and implementation are presented in this section. The need for more than one CTM in the GTCMS is explained. Each of the CTMs that comprise the GTCMS is described in detail. Research that must be carried out to support GTCMS development is described. Guidance on the suggested priority for doing this research is also given.

2.1 NEED FOR THREE CHEMICAL TRANSPORT MODELS IN THE GLOBAL TROPOSPHERIC CHEMISTRY MODELING SYSTEM

The RITS can be divided into three classes, based on their atmospheric lifetimes, their spatial variability in the troposphere, and their sensitivity to cloud processes and precipitation scavenging:

1. short-lived species--CO, NO_x, O_3, NMHCs; lifetimes of days to weeks (CO is longer); high spatial variability; moderately sensitive to clouds and precipitation

2. long-lived species--CH_4, N_2O, chlorofluorocarbons (CFCs); lifetimes of years; low spatial variability; insensitive to clouds and precipitation

3. aerosols--H_2SO_4, $(NH_4)_2SO_4$, NH_4NO_3, MSA, elemental carbon; lifetimes of days to weeks; high spatial variability; high sensitivity to clouds and precipitation.

As discussed below, modeling these different classes of species involves different emphases. A significant recommendation by the research team is that a suite of three interrelated CTMs is needed in the GTCMS to address the distributions of all RITS.

Reaction rates of the short-lived species are comparable or even faster than their rates of vertical mixing and horizontal dispersion in the troposphere. Thus, the model time step will be controlled by the chemical time scale for these species. Concentrations of these species are sensitive to aqueous-phase chemical reactions (Lelieveld and Crutzen 1991) and transport by convective clouds (Brost et al. 1988), so cloud processes are important, though not as much as for aerosol species. Because the short-lived species are photochemically active, the diurnal variation in their concentrations may

be large. Spatial distributions of the short-lived species are strongly affected by the locations of their (or their precursors) major emissions sources. Hence, the CTM for short-lived species must resolve significant horizontal and vertical gradients in concentrations. Photochemical reactions among the short-lived species give rise to the oxidants; namely, OH, HO_2, H_2O_2, and oxygenated organic radicals, that also interact with the long-lived species. Concentrations of these oxidant species calculated by the short-lived species CTM would be used as inputs to the long-lived and aerosol CTMs.

The long-lived species react relatively slowly in the troposphere. CFCs and N_2O are essentially unreactive in the troposphere; CH_4 is slowly reactive. Thus, CH_4 is the only radiatively active, long-lived species of concern. For these species, the rates of vertical mixing and horizontal dispersion are considerably greater than the rate of chemical transformation within the troposphere. Thus, the duration of the model time step may be limited by the advective rather than the chemical time scale. By being rapidly well mixed in the troposphere, the vertical and horizontal resolutions of the CTM for the long-lived species need not be as fine as for the other species. Most of these species have rather low solubilities and, hence, are not appreciably affected by cloud microphysical processes. As a result, a simplified treatment of clouds that focuses more on convective cloud transport than on scavenging is appropriate for the long-lived species CTM. Furthermore, because of their low reactivity and solubility, dry deposition of these species to the surface is not considered to be an important lower boundary sink. However, the upper boundary, typically set at or above the tropopause, needs special attention because the stratosphere may act as a source or sink for these species. CO_2 is not included among the long-lived species because it is not at all reactive in the troposphere. The tropospheric chemical reactions giving rise to CO_2 (oxidation of CO, CH_4, and NMHCs) have only a minor effect on its tropospheric abundance. Also, because of the importance of the oceanic reservoir of CO_2 and changes in the reservoir resulting from changes in ocean circulations associated with climate changes, a stand-alone atmospheric model for CO_2 would be of limited value.

Aerosols need to be treated separately from either the short-lived or long-lived gaseous species because of the need to resolve their size-dependent

composition, their interaction with cloud microphysical processes, and their heterogeneous chemical reactions. Unlike gas-phase species, which are affected by but have little effect on clouds, aerosols significantly affect cloud microphysics. Within the troposphere, photochemical production of aerosols through the oxidation of precursor gaseous species, such as SO_2 and DMS, and neutralization reactions of acidic species (H_2SO_4 and HNO_3) by NH_3 occur at rates comparable to reactions between the short-lived species. Interactions between aerosol and cloud and raindrops are so highly size dependent that correct treatment requires simulating the size distribution of these classes of particles. Detailed information is needed on aerosol size-dependent concentration and composition to properly calculate the radiative transfer properties of aerosols and clouds in the troposphere.

While it is conceptually possible to devise a single CTM that handles all classes of RITS, the development of separate CTMs in the GTCMS for each class is recommended. In practice, the three CTMs would utilize different modules in the common computational framework provided by the GTCMS. As noted elsewhere herein, the Models-3 system (Dennis and Novak 1991) could provide that framework. Through a common computational framework (i.e., common input/output formats, grid structure, numerical solvers, and modular design), the three CTMs can be kept internally and externally consistent.

2.2 DESIGN DETAILS FOR THE THREE CLASSES OF CHEMICAL TRANSPORT MODELS

A basic premise of the design of the GTCMS is that it will be modular (i.e., to the maximum extent possible, the various processes to be simulated by the model will be treated in separate sections of code, herein called modules). The exchanges of information among modules will need to be well defined by the modeling system. The Models-3 framework for modular code development and execution, currently being developed by the EPA's Atmospheric Research and Exposure Assessment Laboratory (AREAL) for use with air quality models, has the potential of also serving as the framework for the GTCMS.

The various inputs required and outputs provided by each module are listed for the three CTMs in Table 2.1. The table is partitioned into broad categories of key processes (e.g., chemistry, dynamics, etc.) with which key subprocesses are listed (e.g., gas-phase chemistry, aqueous-phase chemistry,

TABLE 2.1. Input and Output Variables by Module for the Three Classes of Chemical Transport Models in the Global Tropospheric Chemistry Modeling System

Module/ Class	Input Field	Input Source	Output Field
CHEMISTRY			
Gas phase: Calculates changes in species concentrations from gas-phase chemical reactions			
Short-lived CTM	Species concs.	Primary	Species concs.
	Rate constants	Model parameters	Production rates
	Actinic flux	Primary	
	Water vapor	Meteorological model	
	Temperature	Meteorological model	
Long-lived CTM	Hydroxyl (OH)	Short-lived CTM	CH_4 concs.
	Rate constants	Model parameters	
	Temperature	Meteorological model	
	CH_4 concs.	Primary	
Aerosol CTM	Hydroxyl (OH)	Short-lived CTM	Species concs.
	Rate constants	Model parameter	Production rates
	Species concs.	Primary	
	Temperature	Meteorological model	
Aqueous phase: Calculates changes in species concentrations from aqueous-phase chemical reactions			
Short-lived CTM	Gas-phase spec. concs.	Primary	Gas-phase spec. concs. Aqu.-phase spec. concs.
	Mass transfer coeff.	Model parameters	
	Cloud water	Primary	
	Cloud droplet number	Primary	
	Rain water	Primary	
	Rain drop number	Primary	
	Aqu. rate constants	Model parameter	
	Henry's law consts.	Model parameter	
	Equilibrium consts.	Model parameter	
	Temperature	Meteorological model	
Long-lived CTM	None	None	None

TABLE 2.1. (contd)

Module/ Class	Input Field	Input Source	Output Field
Aerosol CTM	H_2O_2 and O_3 concs.	Short-lived CTM	Gas-phase S spec. concs.
	Gas-phase S spec. concs.	Primary	Aqu.-phase S spec. concs.
	Mass transfer coefficients	Model parameter	
	Cloud water	Primary	
	Cloud droplet number	Primary	
	Rain water	Primary	
	Rain drop number	Primary	
	Aqu. S rate consts.	Model parameter	
	Henry's law consts.	Model parameter	
	Equilibrium consts.	Model parameter	
	Temperature	Meteorological model	
Aqueous size dependence: With bulk approach, variations in aqueous-phase species concentrations with cloud or rain droplet size are not treated			
Short-lived CTM	Mean cloud droplet radius	Model parameter (or primary)	Bulk aqueous-phase species concs.
Long-lived CTM	None	None	None
Aerosol CTM	Mean rain drop radius	Model parameter (or primary)	
Subgrid: Calculates species concentrations in point source or urban plumes until they become fully mixed in model grid cells			
Short-lived CTM	Point source emissions	Emissions inventory/ model	Modified species Concs. at nodes
	Subgrid point source locations	Emissions inventory/ model	
	Wind components	Meteorological model	
	Mixed layer depth	Meteorological model	
	Species concs. at nodes	Primary	
Long-lived CTM		(no module needed)	
Aerosol CTM		(no module needed)	

TABLE 2.1. (contd)

Module/ Class	Input Field	Input Source	Output Field
Aerosols: Calculates changes in aerosol chemical composition and physical size distribution from condensation and evaporation of gases, coagulation, new particle formation, and heterogeneous chemical reactions			
Short-lived CTM	Aerosol mass	Primary	Aerosol mass
	Aerosol composition	Primary	Aerosol composition
	Aerosol size distrib. params.	Model parameters	Volatile gases conc.
	Volatile gas conc.	Primary	
	Nonvolatile species production rates (H_2SO_4, etc.)	Primary	
	Aerosol emissions (mass and composition)	Emissions inventory/model	
	Temperature	Meteorological model	
	Pressure	Meteorological model	
	Relative humidity	Meteorological model	
Long-lived CTM	None	None	None
Aerosol CTM	Aerosol number	Primary	Aerosol number
	Aerosol mass	Primary	Aerosol mass
	Aerosol composition	Primary	Aerosol composition
	Aerosol size distrib. params.	Model parameters	Volatile gases conc.
	Volatile and nonvolatile gas concentrations	Primary	
	Aerosol emissions (number, mass and composition)	Emissions inventory/model	
	Temperature	Meteorological model	
	Pressure	Meteorological model	
	Relative humidity	Meteorological model	
DYNAMICS			
Transport: Calculates changes in species concentrations from advection by resolved winds and turbulent motions			
Short-lived CTM	u,v,w wind components	Meteorological model	Species concs.
Long-lived CTM	Species concs.	Primary	Species concs.

TABLE 2.1. (contd)

Module/ Class	Input Field	Input Source	Output Field
Aerosol CTM	Eddy diffusivities Boundary conditions Transport time step	Primary Model input Model parameter	Species concs.
Boundary layer/turbulence: Calculates turbulent transport parameters (e.g., eddy diffusivities) from other meteorological parameters			
Short-lived CTM	Temperature	Meteorological model	Vertical mass transfer rates
Long-lived CTM	Pressure	Meteorological model	Vertical mass transfer rates
Aerosol CTM	u,v,w wind components Moisture Stability Terrain Land use Surface roughness	Meteorological model Meteorological model Meteorological model Fixed Fixed Fixed	Vertical mass transfer rates
Stratosphere/troposphere exchange: Calculates changes in species concentrations from transport between the troposphere and stratosphere			
Short-lived CTM	Climatology of strat. O_3 and NO_y concs. Julian day	Model parameters Primary	Strat./tropo. flux of O_3 and NO_y
Long-lived CTM	Strat. O_3 and NO_y concs.	Model parameters	Strat./tropo. flux of O_3 and NO_y
Aerosol CTM	Strat. O_3 and NO_y concs.	Model parameters	Strat./tropo. flux of O_3 and NO_y

CLOUD PROCESSES

Microphysics: Calculates microphysical parameters (cloud water concentration, precipitation rate, ...) from other meteorological parameters for use by other modules. Calculates subgrid spatial variations of cloud parameters in stratiform and shallow convective clouds for use by other modules

TABLE 2.1. (contd)

Module/ Class	Input Field	Input Source	Output Field
Short-lived CTM	Water vapor	Meteorological model	Cloud water mass
	Temperature	Meteorological model	Precipitation rate
	Pressure	Meteorological model	Subgrid cloud fraction
	Vertical velocity	Meteorological model	
	Cloud water mass	Primary	
	Precipitation rate	Primary	
Long-lived CTM	None	None	None
Aerosol CTM	Water vapor	Meteorological model	Mean cloud water mass and drop number
	Temperature	Meteorological model	Mean cloud ice mass and ice crystal number
	Pressure	Meteorological model	Mean precipitation
	Vertical velocity	Meteorological model	Variance of cloud water mass and drop number
	Subgrid vert. vel.	Meteorological model	Variance of cloud ice mass and ice crystal number
	Cloud drop. nuc. rate	Primary	Variance of precipitation rate
	Mean cloud water mass, number	Primary	Subgrid cloud fraction
	Mean cloud ice mass, number	Primary	
	Mean rain water mass, number	Primary	
	Mean precipitation rate	Primary	
	Cloud water variance	Primary	
	Cloud drop number var.	Primary	
	Cloud ice variance	Primary	
	Cloud ice number var.	Primary	
	Precipitation variance	Primary	

Scavenging: Calculates changes in species concentrations from interactions with cloud and precipitation particles and precipitation fallout

Short-lived CTM	Cloud water	Primary	Soluble gases conc.
	Precipitation	Primary	Aerosol mass
	Subgrid cloud fraction	Primary	Aerosol composition
	Gas concentrations	Primary	
	Rain water	Primary	
	Rain drop number	Primary	
	Aerosol mass	Primary	
	Aerosol composition	Primary	
	Aerosol size distrib. params.	Model parameters	
	Dissolved gas conc.	Primary	
	Dissolved aero. mass	Primary	
	Dissolved aero. comp.	Primary	

TABLE 2.1. (contd)

Module/ Class	Input Field	Input Source	Output Field
Long-lived CTM	None	None	None
Aerosol CTM	Cloud water	Primary	Soluble gas concs.
	Cloud drop number	Primary	Aerosol number, mass, composition
	Precipitation	Primary	
	Subgrid cloud fraction	Primary	
	Gas concentrations	Primary	
	Aerosol number, mass, composition	Primary	
	Aerosol size dist. params.	Model parameters	
	Dissolved gas conc.	Primary	
	Dissolved aero. number, mass, composition	Primary	
Activation: Calculates activation of aerosols to cloud droplets during cloud formation			
Short-lived CTM	Aerosol mass	Primary	Aerosol mass
	Aerosol composition	Primary	Aerosol composition
	Aerosol size distrib. params.	Model parameters	Cloud drop nucleation rate
	Vertical velocity	Meteorological model	
	Temperature	Meteorological model	
	Pressure	Meteorological model	
Long-lived CTM	None	None	None
Aerosol CTM	Aerosol number	Primary	Aerosol number
	Aerosol mass	Primary	Aerosol mass
	Aerosol composition	Primary	Aerosol composition
	Aerosol size distrib. params.	Model parameters	Cloud drop nucleation rate
	Vertical velocity	Meteorological model	
	Subgrid vert. vel.	Meteorological model	
	Temperature	Meteorological model	
	Pressure	Meteorological model	
Convective clouds: Calculates changes in species concentrations from vertical transport by convective clouds			
Short-lived CTM	Cumulus mass flux	Meteorological model	Gas concentrations
	Gas concentrations	Primary	Aerosol mass

TABLE 2.1. (contd)

Module/ Class	Input Field	Input Source	Output Field
	Aerosol mass	Primary	Aerosol composition
	Aerosol composition	Primary	
Long-lived CTM	Cumulus mass flux	Meteorological model	Gas concentrations
	Gas concentrations	Primary	
Aerosol CTM	Cumulus mass flux	Meteorological model	Gas concentrations
	Gas concentrations	Primary	Aerosol number
	Aerosol number	Primary	Aerosol mass
	Aerosol mass	Primary	Aerosol composition
	Aerosol composition	Primary	

SURFACE EXCHANGE

Calculates changes in species concentrations from exchange between the atmosphere and land/ocean surfaces

Short-lived CTM	Gas-phase species emissions rates	Emissions inventory/ model	Gas-phase species net removal rate at surface
	Gas-phase species concs.	Primary	Aerosol net removal rate at surface
	Canopy resistance by species and land use	Model parameters	
	Surface wind speed	Meteorological model	
	Surface temperature	Meteorological model	
	Stability	Meteorological model	
	Surface roughness and cover	Fixed	
Long-lived CTM	Gas-phase species emissions rates	Emissions inventory/ model	Gas-phase species net removal rate at surface
	Gas-phase species conc.	Primary	
	Deposition velocities by species	Model parameters	
Aerosol CTM	Gas-phase species emissions rates	Emissions inventory/ model	Gas-phase species net removal rate at surface
	Gas-phase species conc.	Primary	Aerosol net removal rate at surface
	Aerosol emissions rate	Emissions inventory/ model	
	Aerosol concentration	Primary	

TABLE 2.1. (contd)

Module/ Class	Input Field	Input Source	Output Field
	Canopy resistance by species and land use	Model parameters	
	Surface wind speed	Meteorological model	
	Surface temperature	Meteorological model	
	Stability	Meteorological model	
	Surface roughness and cover	Fixed	
ACTINIC FLUX			
Radiative transfer: Calculates actinic fluxes and photolysis rates and effect of subgrid spatial variability of clouds on actinic fluxes and photolysis rates			
Short-lived CTM	Latitude and longitude	Primary	Actinic flux
	Local time	On-line calculation	
	Altitude	Primary	
	RITS concentrations	Primary	
	Subgrid cloud fraction	Primary	
	Cloud water mass	Primary	
	Cloud droplet number	Primary	
Long-lived CTM	Latitude and longitude	Primary	Actinic flux
	Local time	On-line calculation	
	Altitude	Primary	
Aerosol CTM	Latitude and longitude	Primary	Actinic flux
	Local time	On-line calculation	
	Altitude	Primary	
	Temperature	Meteorological model	
	Pressure	Meteorological model	
	Relative humidity	Meteorological model	
	RITS concentrations	Primary	
	Subgrid cloud fraction	Primary	
	Cloud water mass and droplet number	Primary	
	Cloud ice mass and ice crystal number	Primary	
	Aerosol mass, number, and composition	Primary	
	Aerosol size dist. parameters	Model parameters	

etc.). Within each subprocess category, the input fields are listed, the source of the input fields is given, and the output fields are listed. Input and output fields are specified for each of the three CTMs. Variables whose source is identified as "primary" are variables that must be defined at all grid nodes within the GTCMS and need to be accessible to a large number of modules. Some variables are provided to the GTCMS from external models or data bases. All meteorological variables (temperature, pressure, relative humidity, wind, etc.) are primary variables that are supplied by the meteorological driver model, which, for example, could be a GCM or hemispheric prognostic model. Emissions inventories and models provide input rates for gaseous and aerosol species in each grid cell for point and area sources of anthropogenic and natural origin. Model inputs include many parameters, such as chemical reaction rate and solubility constants, that are set and remain constant. Also, there are many fixed or prescribed inputs, such as terrain elevation, land use, grid spacings, and time steps. Initial and boundary conditions (species concentrations, physical variables) must be specified for all primary variables internal to the GTCMS.

2.2.1 Short-Lived Species Chemical Transport Model

The short-lived species CTM is designed to simulate global O_3 distributions in the troposphere. To accurately predict O_3 distributions, it is necessary to also simulate NO_x, CO, and hydrocarbon (HC) distributions. Thus, the short-lived species CTM must account for the emission, transport, and photochemical transformations of NO_x, NO_y, various HCs (including CH_4 and CO), and O_3. It is recommended that the short-lived species CTM be used to provide oxidant concentration distribution fields to the long-lived species CTM and possibly, in certain cases, to the aerosol CTM. Consequently, the optimal model reflects the most advanced science that could reasonably be achieved within the 3-year project period. The short-lived species CTM is the core of the GTCMS.

2.2.1.1 Chemistry

To account for the photochemical production and destruction of O_3, a gas-phase chemical mechanism is required that includes basic $NO_x/O_3/HC$ photochemistry. In remote regions, CH_4/CO chemistry must be included; while in

near-continental areas, a multiple lumped HC representation is needed to account for anthropogenic and biogenic emissions. The most important aspect of the multiple lumped HC mechanism is the inclusion of appropriate radical-radical reactions,

$$RO_2 + RO_2 \text{ ---> products.}$$

Radical-radical reactions are not usually significant for the accurate representation of radicals in polluted environments (Seinfeld 1989); however, on a global scale and remote areas in particular, these reactions will be an important sink for odd-oxygen species (Lurmann et al. 1986). Thus, a gas-phase mechanism must be developed that appropriately represents these reactions while keeping in mind that the chemical transformation calculations account for >90% of the total model computational burden. Also, the gas-phase sulfur reactions, DMS to SO_2 and MSA and SO_2 to H_2SO_4, need to be included to treat aerosol processes correctly within the optimal model (Kreidenweis et al. 1991).

In the aqueous phase, the reactions of S(IV) oxidation to S(VI) are included, which requires that $NO_y/O_3/H_2O_2$ aqueous reactions be included (Finlayson-Pitts and Pitts 1986). Aqueous radical reactions of the type proposed by Lelieveld and Crutzen (1991) should be included because these may have a significant effect on O_3 distributions globally. The heterogeneous reaction of N_2O_5 on wetted aerosol particles producing HNO_3 (Dentener and Crutzen 1993) appears important to the global NO_x budget and should also be included. No size dependence to aqueous chemistry or aerosol properties can be justified for inclusion at this time into the short-lived species CTM.

The subgrid treatment of chemical processes would be an important aspect of the short-lived species CTM. In the near future, computational resource limits will prevent global models having the horizontal resolution necessary to fully resolve major NO_x plumes from large urban areas. Within these subgrid plumes, substantial O_3 production can take place that is not translated to the large-scale grid when using only instantaneous dispersion of subgrid sources. The exact form that this subgrid treatment should take is not clear, though it is clear that the instantaneous dispersion treatment is

not be desirable. Development of a plume-in-grid or statistical method (e.g., Sillman et al. 1990) should be a priority research topic.

2.2.1.2 Dynamics

While K-theory approximations will be adequate for the short-lived species CTM, some higher-order dynamics can be incorporated in a relatively straightforward manner by using a turbulent kinetic energy model to generate eddy diffusivity values (e.g., Yamada and Mellor 1975). The K-theory approximation will be adequate in light of the inclusion of a convective cloud parameterization to redistribute material between layers in a nonlocal fashion. For stratosphere/troposphere exchange, a simple boundary condition treatment will not be adequate to accurately simulate the global distribution of O_3.

Thus, the model domain should be extended into the lower stratosphere in which some type of O_3 and NO_y climatology is specified. No chemical calculations would be performed in the lower stratosphere, but a GCM would be used to provide dynamic exchange information for the transport of material across the tropopause. The exchange of mass between the stratosphere and troposphere is currently not very well understood (Slinn 1993) and should receive significant emphasis in the design of the short-lived species CTM.

2.2.1.3 Cloud Processes

The effects of clouds are an important, though not dominant, factor in the design of the short-lived species CTM. Consequently, bulk treatments of cloud microphysics, scavenging processes, and aerosol activation (nucleation of cloud drops) were chosen. Further, a fractional subgrid cloud treatment is adequate, with cloud convective processes implemented in a manner consistent with the treatment of cumulus convection in the GCM used to provide the meteorological data.

2.2.1.4 Surface Exchange

A surface-exchange model was chosen as a feasible goal for inclusion in the short-lived species CTM. The dry deposition/resistance approach (Wesely 1989) does not allow for the possible two-way exchange of material across the soil/atmosphere or ocean/atmosphere interfaces. Thus, a surface-exchange

model should be developed that can more adequately treat surface-exchange processes. Progress by the agricultural science community on surface-exchange processes should be examined and adapted by the atmospheric science community to the GTCMS.

2.2.1.5 Actinic Flux

The actinic flux calculations for the short-lived species CTM must be more realistic than present-day, look-up-table approaches (e.g., Peterson 1977) but short of on-line radiative flux calculations. The actinic flux will be parameterized to dynamically depend on cloud amounts and will vary with the total cloud fraction and type of cloud within the grid cell. This information would be accessible through look-up tables.

2.2.1.6 Numerics

Time splitting, coupled with either a one-dimensional finite-difference method or a one-dimensional finite-element method on a regular, uniform grid, would suffice for solving the transport equations for this model. The use of an irregular horizontal grid (Mathur et al. 1992) would allow the a priori specification of a grid that would have finer resolution over the continents and coarser grid spacing over remote ocean areas, providing finer resolution only where it is really needed. Although the use of an irregular grid has significant advantages, implementing and testing such a scheme within the 3-year project period is not likely. However, because irregular grids will likely be used in future versions of the model, time splitting with a one-dimensional finite-element method is recommended for the initial version. This would allow for the relatively straightforward modification of the code in future years to use time splitting with irregular two-dimensional elements in the horizontal plane.

Although a new stiff ordinary differential equation (ODE) solver is a high priority for improving the integration of the chemistry rate equations, the quasi-steady-state approximation (QSSA) method (Hesstvedt et al. 1978) is recommended because of its economy and current widespread use. Of course, if a faster, yet accurate, method became available during the development of the model, it should be incorporated.

2.2.2 Long-Lived Species Chemical Transport Model

The long-lived species CTM is capable of simulating the distribution and chemical reactions of RITS over a time scale of decades. The only long-lived species that is chemically reactive in the troposphere is CH_4. The gas-phase chemistry mechanism will require carrying additional reactive species that are not radiatively important, such as OH and certain classes of NMHCs. Non-reactive species, such as the CFCs and N_2O, can be carried in the model as inert tracers.

2.2.2.1 Chemistry

Gas-Phase Reactions. The primary reactions to be explicitly described in the long-lived species CTM are those related to CH_4. It was deemed infeasible to explicitly calculate the OH concentrations necessary for describing the full $CH_4/OH/CO_2$ sequence for model runs on decadal time scales. A necessary alternative is to prescribe the oxidant fields in advance, using output from the short-lived species CTM. In addition to tabulating (or describing in some other readily accessible form) the OH concentrations, information on the sensitivity of OH to CH_4 and other species involved in the CH_4 cycle is needed. These values would have to reflect trends in time (on a seasonal scale) and space.

A significant reduction in computer time would result by not having to carry all the species necessary to calculate OH fields. In addition, the resulting kinetic equations could be evaluated using a nonstiff differential equation integrator, resulting in an additional reduction in computer costs. The use of precalculated OH concentration fields would be the single most important simplification incorporated into the long-lived species CTM so that it could be run in a timely manner for a multitude of planning and policy issues.

Aqueous Reactions. The explicit inclusion of detailed cloud fields, and, hence, of aqueous-phase reactions, would be impossible over the time scales this model would be called on to simulate. If the CH_4 fields respond to cloud processes, those processes would have to be included in some statistical fashion or reflected in the look-up tables for OH (and its associated derivatives).

Aerosols. Aerosols were considered to be significant over decadal time scales only if a feedback process between the chemistry and the atmospheric dynamics were to be simulated. Because this is beyond the scope of the requested design, a simulation of aerosol processes was considered an unnecessary computational burden for this model.

2.2.2.2 Dynamics

Planetary Boundary Layer/Turbulence. A simple first-order turbulence scheme is satisfactory to account for turbulent mixing in the planetary boundary layer.

Stratosphere/Troposphere Exchange. This is an area requiring research, and one where the results could greatly affect simulations of the distribution of the long-lived species in the troposphere. It is unlikely that a detailed mechanistic treatment of stratosphere/troposphere exchange can be included in the GTCMS. Thus, the research should yield a parameterized representation of this process.

2.2.2.3 Cloud Processes

The only cloud process to be explicitly included in the long-lived species CTM is that of convective mixing. A simple formulation, using either mixing coefficients derived from the convective scheme of the GCM that provides the meteorological fields or a decoupled mechanism (Walcek and Taylor 1986 or Berkowitz et al. 1989), would be adequate. The choice of cloud processes parameterizations should be consistent with the approach taken in the GCM that is used to provide the meteorological fields for the GTCMS. This would maintain consistency between the meteorological fields obtained from the GCM and the convective process simulations within the GTCMS.

The omission of an explicit treatment of cloud water may affect the simulated removal of HCHO, which is a soluble species known to play an active role in the CH_4 chemistry. Further research is needed to determine if this omission will have a significant effect.

2.2.2.4 Surface Exchange

Transfer coefficients obtained from the GCM are used to calculate the exchange of CH_4 between the ocean and the atmosphere. CFCs and N_2O can be assumed to have negligible interaction with the surface.

Emissions are a component of the surface boundary condition that is very important to the simulation of the concentrations of the long-lived species. In particular, the geographic and seasonal distribution of CH_4 emissions must be thoroughly specified.

2.2.2.5 Actinic Flux

The use of look-up tables, based on aerosol and trace gas concentrations and meteorological conditions, is recommended for both the magnitude of the actinic flux and its subgrid variability.

2.2.2.6 Numerics

Transport. The Lagrangian method is not recommended for reasons related to computational effort. Although the method of moments is very accurate, the computational effort was not felt to be necessary in light of other simplifications associated with this model. Other than these two caveats, any mass conserving, accurate integration scheme would be acceptable.

Chemistry. Any currently used integration package would be acceptable, though the Gear routine offers the advantage of automatically adjusting the time step to an optimal increment. However, further analysis will be needed to determine how the numerical time steps of a nonstiff chemical integrator would relate to the time steps used in the meteorological driver.

2.2.3 Aerosol Chemical Transport Model

The aerosol CTM will simulate the global distribution of anthropogenic aerosols and their impact on the planetary radiation balance, both by direct scattering and absorption of sunlight, and by serving as CCN. To do this, it is quite clear that the model should treat aerosols of differing composition (e.g., $[NH_4]_2SO_4$, NH_4HSO_4, H_2SO_4, EC, HC, and silicate) that have different optical and cloud nucleating properties and often compete with each other. In addition, the model should distinguish between anthropogenic and natural

aerosols and between aerosols in the fine, accumulation, and coarse modes. The options selected to meet these objectives optimally are discussed below.

2.2.3.1 Chemistry

Aerosol chemistry is not independent of gas-phase chemistry nor, for that matter, is aqueous-phase chemistry independent of gas-phase chemistry. The separation is primarily for conceptual purposes: it is easier to think about these three aspects of chemistry individually rather than all at once. They are, of course, all linked by mass transfer (gases <--> cloud water and gases <--> aerosols). In the model, the chemistry ODE solver might call all three of these chemistry routines to get the rates of change for gaseous, aqueous, and aerosol chemical species. Alternately, the gas and aqueous chemistry might be solved separately from the aerosol chemistry because of differing time scales (stiffness). In this case, the N_2O_5(gas) --> HNO_3(aerosol) reaction might be treated in the gas-phase chemistry module, and the resulting change in HNO_3(aerosol) over the model time step would then be passed to the aerosol module.

The gas- and aqueous-phase chemistry important to the aerosol model involves the transformation of aerosol precursors (SO_2, DMS, organics) to aerosol species. Thus, oxidant species involved in these transformations (OH, HO_2, H_2O_2, O_3) must be available. The appropriate gas-phase mechanism for simulating these oxidants is a hybrid mechanism that treats NO_x, O_3, CO, CH_4, and several lumped HCs over continental regions with significant HC emissions but ignores HCs over other regions to reduce computational costs. Sulfur chemistry (SO_2 and DMS) clearly must be included. Note that because sulfur chemistry has little effect on OH concentrations, multiple simulations with differing SO_2 and particle emissions (but fixed NO_x and HC emissions) could be performed by saving the OH from the first simulation and reusing it in subsequent simulations. A much simpler, though less realistic, alternative for obtaining OH is to parameterize it as a function of several variables, such as actinic flux, humidity, temperature, CO, O_3, and NO_y (Spivakovsky et al. 1990). Further evaluation of this approach versus the direct computation of OH is needed. NH_3 is very important because it determines whether sulfate is present as H_2SO_4, NH_4HSO_4, or $(NH_4)_2SO_4$. Because NH_3 emissions are poorly known, it is not clear whether direct computation or an

empirical parameterization of its concentration would produce better results. An approach following Sillman (1991) is recommended for the time integration of the full chemistry; if OH is prescribed, the chemical system is no longer stiff and a simple, nonstiff scheme is adequate.

Aqueous-phase chemical reactions could be limited to the reactions of dissolved SO_2 with H_2O_2 and O_3. Other reactions, such as SO_2 oxidation by trace metals and OH, and aqueous-phase H_2O_2 production are currently felt to be of secondary importance. The main difficulty with the SO_2-trace metal reaction is specifying trace metal concentrations. Simulating aqueous free radical chemistry is computationally expensive. Solution chemistry involving soluble gases (NH_3, HNO_3) and aerosol species (NH_4, NO_3, etc.) should also be included. In addition to gas- and aqueous-phase chemistry, some heterogeneous reactions involving gases and aerosols should be considered. Examples are the oxidation of SO_2 to sulfate in deliquescent sea-salt particles (Chameides and Stelson 1992) and the reaction of N_2O_5 on wetted aerosol particles producing HNO_3. Although evidence is accumulating that aqueous-phase chemistry depends to some extent on cloud droplet size, it was not felt that this has a primary effect on aerosol concentrations and size distributions, and including it in the model is not justified because of the added computational burden.

Because aerosol light-scattering and droplet-nucleating properties are strongly size dependent, some information on the aerosol size distribution must be included in the model to calculate the radiative effects of aerosols. The method of moments offers an efficient means of representing the size distribution of each aerosol moment without the very large computational burden of explicitly treating the size distribution. Each mode of the size distribution is expressed in terms of an analytic function (log-normal is generally used) whose parameters can be related to just a few moments of the aerosol size distribution. The parameters of the log-normal distribution (total number, geometric mean radius, and geometric standard deviation) can be related to any three integer moments of the size distribution. This approach is physically justified by the fact that observed aerosol size distributions can often be accurately approximated by combinations of log-normal functions (Whitby 1978). Considerable development of the method of moments was done by

Giorgi (1986) and Whitby et al. (1991). The method is currently being used in EPA's Regional Particulate Model.

Aerosol growth processes, such as coagulation, vapor condensation, surface reactions, and aqueous chemical reactions in cloud drops followed by drop evaporation, lead to aerosol particles that are mixtures of several chemical species. Such internal mixtures can have optical and nucleating properties that are quite different from external mixtures of single-component aerosols, so the degree of internal mixing of the aerosol components is an important issue. There are three strategies for modeling the aerosol composition. The external mixing strategy assumes that individual particles are composed of a single chemical species (i.e., sulfate and carbon are found in different particles). The internal mixing strategy assumes that, within a given mode, each particle is the same mixture of the chemical components of the mode (i.e., each particle is 70% sulfate and 30% carbon). Both of these strategies are easily implemented but neither is realistic, though the internal mixing strategy is fairly good for the accumulation mode. The partial mixing strategy recognizes that, at a given size, particles with many different chemical mixtures exist. This strategy is most appropriate for the aerosol model; however, it is not straightforward and additional research is needed to implement it.

2.2.3.2 Dynamics

Turbulent mixing should be treated in a manner consistent with the treatment in the meteorological model. First-order K-theory is adequate for the optimal model. Tropospheric-stratospheric exchange is of secondary importance for aerosols and can be treated as an inflow-outflow boundary with prescribed stratospheric concentrations.

2.2.3.3 Cloud Processes

Cloud microphysical processes play a critical role in processing aerosols and in determining indirect radiative impacts of aerosols. The treatment of cloud microphysics considered most attractive for this application is the method of moments, in which a few moments of the cloud particle-size distribution are predicted (Clark 1976; Clark and Hall 1983; Ziegler 1985; Verlinde et al. 1990). As a minimum, the cloud particle number and volume would be

predicted, which permits a treatment of the influence of aerosols on cloud particle size and provides adequate information on cloud radiative properties. Cloud microphysical processes and their impact on each moment would be treated by assuming idealized cloud particle-size distributions. Cloud ice should be distinguished from cloud liquid water because it affects aerosols and gaseous precursors in a much different manner. It may be possible to ignore cloud ice in the GTCMS (though not in the meteorological driver) because cloud ice generally has a much smaller impact than cloud water on aerosols and gases.

The nucleation of cloud droplets on aerosols (activation) should be treated by accounting for the influence of preferential nucleation on all moments of each aerosol mode. The competition between different aerosol components and different aerosol modes must be represented.

Given that the method of moments is used to represent both aerosols and cloud particles, it is natural to use the method of moments to treat aerosol scavenging by clouds. Analytic integrations over size distributions of aerosol and precipitation would be performed using assumed idealized size distributions.

When aerosols enter a cloud, two different aerosol populations are formed: those in the interstitial air and those in cloud droplets. Each of these have separate size distributions and are affected by different processes (e.g., aqueous chemical reactions and cloud droplet coalescence affect those in cloud droplets). A methodology for treating these two populations and combining them when a cloud evaporates needs to be developed.

A realistic treatment of subgrid variability in clouds and cloud processes is critical for global chemistry models. Two options are considered. Subgrid variability could be expressed in terms of idealized probability distributions of each cloud variable, with several moments of the probability distribution (e.g., mean and variance) predicted by integrating all microphysical processes over the probability distributions of the cloud variables. Although this approach is likely to be the state of science within 5 years, it relies on estimates of subgrid variability in vertical velocity, which is a very challenging task. The optimal model would, instead, simply express subgrid variability in terms of a cloud fraction, with the clear and cloudy

fractions assumed to be homogeneous. Although this is a crude representation of subgrid variability, it captures much of the variability and is much easier to parameterize than a full probability distribution.

Vertical transport and scavenging of chemical species by convective clouds should be treated in a manner consistent with the treatment of cumulus convection in the meteorology model. A single-plume updraft/downdraft version of the mass flux scheme of Arakawa and Schubert (1974) is optimal, for it provides a straightforward means of treating transport and scavenging of all species without the high computational cost of the full spectral Arakawa and Schubert (1974) parameterization.

2.2.3.4 Surface Exchange

Global emissions inventories are needed for each primary particulate aerosol type and all aerosol precursors. For primary particles, emissions of smoke and soot from industrial sources, transportation, and biomass burning (natural and agricultural) and suspension of dust are needed. Gaseous precursors to aerosols include SO_2, DMS, NH_3, NO_x, and organics. For natural emissions, dependence on wind speed and stability should be represented using resistance formulations. For anthropogenic emissions, seasonal dependence should be accounted for. The treatment of surface exchange involving emission products should also use the resistance formulation.

2.2.3.5 Actinic Flux

For the purpose of simulating OH, actinic flux is needed for photochemical mechanisms. The look-up-table approach should be augmented by an on-line treatment of the influence of clouds on actinic flux. This can be accomplished simply by using a two-stream radiation model with the cloud optical properties and subgrid variability to determine the ratio of the cloudy sky actinic flux to that for clear-sky conditions. The ratio should not vary much with wavelength, so that the same ratio can be applied to the clear-sky look-up table for all photochemical absorption bands.

2.2.3.6 Numerics

Transport should be calculated with a one-dimensional finite-difference or finite-element algorithm. The added complexity of two-dimensional or

method of moments is not warranted; a fixed irregular grid system would be optimal because higher resolution could be given to source regions with little additional computational burden. However, additional software development is required for interpolating meteorological, emissions, and land-use data from a regular to irregular grid and for analysis of simulation results. Unless the irregular grid is selected for another model (e.g., short-lived gases), this development may not be justified.

2.3 UNCERTAINTY/SENSITIVITY

Model uncertainty has several components. Uncertainty arises from the fact that the mathematical descriptions of physical and chemical processes in the model only approximate real-world processes. Additional uncertainty arises from the spatial resolution of the model (100 to 500 km in a GTCMS), which leaves smaller-scale processes unresolved. The techniques described below apply primarily to quantifying uncertainty from indeterminant model parameters (e.g., chemical reaction rates), initial conditions, and boundary conditions. Sensitivity analyses quantify the response of model output (e.g., a predicted concentration) to variations in a particular model parameter or input variable about a base state. Note that while model uncertainty is ultimately of most interest, calculation of model sensitivity is of value and is easier to do. Also note that performing a complete sensitivity of a three-dimensional atmospheric chemistry model is a gigantic computational task. At best, uncertainty is dealt with through an analysis of the sensitivity of the model to various inputs and model parameters. Various approaches for uncertainty and sensitivity analyses are briefly discussed below.

The Monte Carlo method, used for both sensitivity and uncertainty analyses, involves doing repeated model simulations with different values of an uncertain model parameter or initial or boundary condition. The sensitivity of a model prediction (e.g., O_3 concentration) to a model parameter is simply the change of the predicted value relative to the change in the model parameter between two different simulations. To obtain the uncertainty of a model prediction, probability distributions for all model parameters of concern must be specified. (This could be simply a normal distribution with

specified mean and standard deviation.) The mean and standard deviation for the predicted values give a measure of model uncertainty.

The direct decoupled method is a sensitivity analysis technique that has been applied primarily to photochemical mechanisms (Dunker 1984). Additional code is added to the model so that it predicts both concentrations of various species and the sensitivities of these concentrations to one or more model parameters or initial or boundary conditions. The similarity of the ODEs for the sensitivity coefficients and those for the species concentrations results in considerable computational savings during solution of the equations.

While the Monte Carlo and direct decoupled methods compute the sensitivity of all model predictions (every species at every grid point) to a single model parameter, the adjoint method computes the sensitivity of a single prediction (e.g., a spatial and temporal average of the O_3 concentration) to multiple model parameters (Hall et al. 1982). The method involves construction and solution of a second model, called the adjoint, that predicts the sensitivity coefficients. This method is receiving much interest in four-dimensional data assimilation, and methods for automatic generation of adjoint codes are being developed. Monte Carlo analyses are impractical for the computationally intensive short-lived species CTM. An adjoint technique, implemented with automatic differentiation (Griewank and Corliss 1991), is a very attractive option if it could be implemented within the project time period.

The stochastic variables method is a new and promising technique for uncertainty analysis. Stochastic variables are a set of orthogonal random variables. The probability distribution for each uncertain model parameter and initial condition is expressed as a series of one or more stochastic variables. The resulting model predictions then become a series of these stochastic variables (rather than just a number), and this series represents the probability distribution of the predicted value.

Automatic differentiation is a major breakthrough in scientific computing (see Griewank and Corliss 1991), is a fundamentally new technology for reliably computing derivatives of large computer codes many times faster compared to current approaches, and is based on a completely different

approach than the finite-difference approximation of derivatives or symbolic differentiation. This emerging technology has been recently incorporated in general-purpose automatic differentiation software packages, such as ADIFOR and ADOL-C developed at Argonne National Laboratory. Given a Fortran program describing how "dependent" variables y1,y2,...,yn are obtained from "independent" variables x1,x2,...,xm, ADIFOR automatically generates a Fortran code for the computation of the derivatives of the dependent variables with respect to the independent variables. ADOL-C is a similar tool for automatic differentiation of programs written in C or C++.

It is clear that tools for integrating uncertainty and sensitivity analyses into complex numerical models now exist and are rapidly being improved. It is recommended that these state-of-science methods be incorporated into the numerical codes for the GTCMS as it is developed.

2.4 GENERAL DISCUSSION OF RESEARCH NEEDS AND ACTIVITIES

Each class of CTM in the GTCMS represents a model development goal. Many of the options that define a chosen CTM do not exist; however, for nearly every module, at least one option is available. The options that are available define the starting point for developing the GTCMS. By analyzing what research was needed to move from the current existing option to the chosen option for each module, several research areas were identified that could be addressed within a 3-year time frame and that could significantly improve the performance of the GTCMS. Very brief descriptions of the research activities in each research area were developed. Table 2.2 gives the title of those research activities and associates the title with an abbreviated title that is used in other tables. A more detailed description of these research activities is given in Appendix C.

The design and construction of the GTCMS will be a complex undertaking that will ultimately require scientific and systems advances along many pathways that must be integrated and coordinated to make the GTCMS a reality.

2.4.1 Chemistry

Many of the research activities associated with the chemistry modules are interrelated and most are critical for the development of a scientifically

TABLE 2.2. Titles of Research Activities Identified by Participants at the Second Workshop as Needed for the Development of the Global Tropospheric Chemistry Modeling System

Module	Abbreviation	Research Activity
Clouds, Aerosols, Radiation		
Nucleation	Mixed aerosols	Parameterize nucleation on mixed aerosols
Subgrid Cloud Processes	Subgrid variability	Develop treatment of subgrid variability in clouds and cloud processes
Aerosols	Moments/species	Develop species-resolved moments of aerosol-size distributions
	Evaluate moments	Evaluate species-resolved moments method
	Primary emissions	Emissions of primary particulate and aerosol precursors
Scavenging	Aerosols/clouds	Processing of aerosols by clouds using moments method
Actinic Flux	Update J-values	Update actinic flux J-value look-up tables
Chemistry		
Gas Phase	$NO_x/O_3/HC$	Develop appropriate $NO_x/O_3/HC$ chemical mechanism for species CTM
	Grid res/NO_x	Evaluate effect of grid resolution on NO_x cycling and O_3 production
	Chem/aerosol	Develop chemical module for aerosol CTM
Emissions	Emissions inven	Develop emissions inventories required for the GTCMS
Aqueous Reactions	Aerosols/O_3	Evaluate effect of aerosols on tropospheric O_3
	Clouds/O_3	Evaluate effects of clouds on tropospheric O_3
Clouds	Clouds/CH_4/CO	Role of clouds in CH_4/CO cycle vis-a-viz HCHO and organic acids
Other Chemistry	Interfaces	Develop interface between short-lived, long-lived, and aerosol CTMs
	OH fields	Evaluate use of short-lived species CTM in providing OH fields for use in long-lived and aerosol CTMs
	DMS chemistry	Evaluate adequacy of gas-phase oxidation mechanisms of DMS and resulting production of CCN
Meteorology and Numerics		
Dynamics	Strat/tropo exch	Stratosphere/troposphere exchange
	Nonlocal	Planetary boundary layer/turbulence nonlocal closure
Surface Exchange	Hydro/met synth	Synthesize into consistent framework models for hydrology, surface exchange, emissions, and deposition
Transport/Grid System	Irreg grid chem	Develop irregular grid method for chemistry
Numerics	Fast integrator	Develop numerically fast chemistry integrator
Uncertainty	Autodiff demo	Carry out demonstration of autodifferentiation using test model or module

credible GTCMS. Many of the activities could be grouped into a larger single activity with smaller tasks within this larger project. Considering that the chemical mechanisms for the GTCMS will be a vitally important part of the entire system and that, by definition, there should be a high degree of chemical consistency between the classes of CTMs, a composite project may be desirable.

2.4.1.1 Gas Phase

A primary research task will be the development of a gas-phase chemical mechanism that will be appropriate for the short-lived species CTM. This must include an appropriate, but not excessive, number of lumped HC categories and associated secondary organic products. Particularly important for a global model will be the proper representation of radical-radical reactions in low-NO_x regimes. Further, the chemical module must be designed for flexibility, so that future advances can be readily incorporated. An excellent way to begin the chemical mechanism construction process would be to conduct a workshop that would bring together experimental atmospheric chemists and modelers to compile the latest understanding of gas-phase chemical processes that are important on the global scale.

One portion of the overall task of constructing an appropriate gas-phase chemical mechanism will be to evaluate the current understanding of DMS tropospheric chemistry. This will include the relative production of SO_2 and MSA from DMS and the resulting production of CCN. This activity must be coordinated with the mechanism development effort and, thus, must begin early in the design process.

For the aerosol CTM, sulfur chemistry will be important but also sources and precursors of aerosols, such as dust, sea salt, biomass-burning emissions, HNO_3, and NH_4. As a consequence, a gas- and aqueous-phase chemical module will have to be developed for the aerosol CTM that takes these other aerosol sources and precursors into account.

Within the GTCMS, the long-lived species and aerosol CTMs act as engineering models in which the short-lived species CTM would provide oxidant fields for detailed scenario analyses by the engineering models. This concept would be similar to the design of the EPA's Regional Acid Deposition/

Engineering Models. Consequently, it is recommended that a project be initiated to evaluate the feasibility of using OH, O_3, and H_2O_2 fields from the short-lived species CTM as input into the long-lived species and aerosol CTMs. If the concept proves feasible, then an interface must be designed between the short-lived species CTM and the other CTMs. The interface should allow for both a "screening mode" (coarse temporal and spatial resolution) transfer of fields and a "detailed mode" (fine temporal and spatial resolution).

2.4.1.2 Subgrid

On the regional scale, much recent work has concerned itself with the effect of subgrid chemical effects. Within the typical 80- x 80-km regional model grid cell, separate plumes from major point or area sources are not resolved. Consequently, the production/destruction of O_3 that would take place in these plumes is not adequately resolved. A research activity should be initiated to investigate the impact of even coarser horizontal grid resolutions that will be required for global-scale simulations and, if possible, to recommend parameterizations to account for these subgrid effects in the global scale.

2.4.1.3 Aqueous Phase

Two related research activities were identified that deal with the effect of cloud processes and aerosols on the production/destruction of O_3 in the troposphere. Recent research has indicated that O_3 production/destruction may be significantly affected by heterogeneous chemical processes in the troposphere. Aqueous-phase processes have been shown to yield a net destruction of O_3 in the presence of clouds (Lelieveld and Crutzen 1991), and sea-salt particles in the marine boundary layer have been implicated as possible sites of heterogeneous O_3 destruction (Chameides and Stelson 1992). These processes should be investigated further to determine their significance for global-scale O_3 distributions.

A similar study should be carried out to evaluate the chemical impact of neglecting cloud processes in the long-lived species CTM. To a first approximation, the neglect of cloud processes is justified because of the relatively small direct effect that aqueous-phase processes have on CH_4 and CO

concentrations. However, HCHO, an intermediate product in the oxidation of CH_4 to CO, is relatively soluble in the aqueous phase and its distribution will probably be significantly affected by the presence or absence of clouds. An investigation should be performed to determine the impact of these processes on the long-lived species predictions.

2.4.1.4 Aerosols

Considerable research is needed to develop the aerosol CTM. The method of moments needs to be developed for multiple aerosol-size modes and aerosol compositions, with treatments of aerosol formation, condensation, coagulation, deliquescence, cloud-drop nucleation, resuspension, scavenging, and settling. Nucleation of drops on aerosols of varying size and composition requires particular attention. Global emissions inventories are needed for primary particles and aerosol precursors. The simulation of aerosols by the model must be evaluated by comparisons with observations.

2.4.2 Dynamics

2.4.2.1 Stratosphere/Troposphere Exchange

The justification for a clear understanding of this exchange process seems evident; it controls inflow and outflow at the top of the entire GTCMS domain. Unfortunately, not only the magnitude but also the direction of this exchange process are uncertain. Part of the difficulty in assigning even a climatological/global-scale average value to this exchange process is that it now appears to have a systematic geographic variability, with net upflow in the tropics and downflow associated with tropopause folding in midlatitudes and upflow in polar latitudes (Hoerling et al. 1993). Additional discussion of our lack of understanding of this process is found in Slinn (1993).

Additional basic research in this area is not recommended for two reasons. First, resolution of these difficulties will not likely occur within the 3-year time frame proposed for development of the GTCMS. Second, the National Aeronautics and Space Administration, National Science Foundation, and various groups in Europe have major programs related to stratospheric chemistry and dynamics. It is strongly recommended that parameterizations be developed that make use of results from these other programs.

2.4.2.2 Planetary Boundary Layer/Turbulence

In classic turbulence exchange (e.g., K-theory), material can be exchanged only among adjacent grids. Nonlocal closure refers to turbulence schemes that describe the transport of material from one layer of a model to another, nonadjacent layer. Work with such nonlocal descriptions using regional-scale models has resulted in significant differences in the final fields, relative to results using more traditional approaches. A key difference is that such methods allow for the transport of material up the concentration gradient; though such processes do occur in nature, they cannot be accounted for in traditional K-theory.

2.4.3 Cloud Processes

Cloud parameterization in GCMs is being addressed by other programs, but certain aspects of clouds may require additional attention for chemistry applications. Subgrid-scale variability in cloud properties and cloud processes has a strong influence on soluble gases and aerosols through deposition, heterogeneous oxidation, activation, scavenging, and resuspension. A physically based treatment of subgrid cloud variability is needed to account for the influence of the variability on soluble gases and aerosols.

2.4.4 Surface Exchange

2.4.4.1 Surface Fluxes

Much work remains to be done before scientists can adequately describe the flux of key chemicals over ocean waters and from developing nations. It is recommended that methods for calculating surface exchange, derived from hydrology, oceanography, urban meteorology, and agriculture, be used. These methodologies, based on vegetation, ocean characteristics, or urban geography, have the potential of making a significant contribution to the GTCMS.

2.4.4.2 Emissions

Emissions, of course, will play a significant role in the design and use of the GTCMS. It is recommended that a research activity be initiated to analyze the emissions requirements of the GTCMS. The emissions needs of each model should include both the species that are necessary for the particular model and the characteristics (e.g., temporal and spatial resolution) of the

inventories that will be important for the model's use. The emissions inventories should cover natural and anthropogenic emissions from land and ocean areas (and aircraft). Species felt to be of particular importance include NH_3, NO_x, SO_x, CH_4, and NMHC. Estimating the source terms for these species over regions difficult to sample (for reasons related to logistics or politics) will be a large but essential undertaking.

2.4.5 Actinic Flux

The look-up tables used to determine photolysis rates need to be revised. Consideration should be given to compatibility with on-line cloud effects on actinic flux.

2.4.6 Numerics

2.4.6.1 Grid System

A key research area that may be resolved within the 3-year time frame of this project is the development of a nested grid system that can interact with the surrounding coarser grid. The uses of a nested grid could be extensive. A few examples of possible applications include enhanced definition of subgrid mixing associated with point sources and simplification of the descriptions necessary for predicted subgrid processes (e.g., cloud fields or other discontinuous physical features).

One difficulty that would have to be addressed prior to including nested grids in the GTCMS is the magnitude of the numerical errors resulting from calculations within the nested grid. This would be a somewhat separate problem from error estimates within just the coarse grid because the boundary conditions within the nested domain would be defined by the surrounding coarser grid. Thus, errors could easily increase within the subdomain.

Related to this question of error propagation was the possibility of designing an integration system that would make the nested grid a "super convergent" system. Super convergence characterizes numerical techniques that result in local accuracy well in excess of what would be expected on a global basis (Fairweather 1978; Oden and Carey 1983). Research on this subject should include this objective as a desirable goal.

2.4.6.2 Numerical Schemes for the Chemistry Integrator

Although many schemes already exist for numerically evaluating the coupled differential equations that describe atmospheric chemical kinetics, few offer any degree of flexibility in their treatment of alternative mechanisms while concurrently maintaining computational efficiency. The development of a fast and flexible chemistry integrator would, therefore, be distinct from an integrator that was fast only for a given mechanism.

Development of this type of chemistry integrator should be pursued. A recent extension of the QSSA by Sillman (1991) results in a large reduction in computational cost and added flexibility. In QSSA, numerical efficiency is increased by lumping together species having similar chemical properties. Sillman's new method, based on reordering species and using an implicit Runge Kutte integration, is suitable for use with stiff equations. Preliminary tests with this extension of the QSSA have resulted in reduced computational complexity and significant reductions in computer resource requirements.

2.5 GUIDANCE ON RESEARCH PRIORITY

Participants at the second workshop applied decision analysis methods (Section 3.4) to rank the research activities listed in Table 2.2. Based on the discounted, weighted average score received by each activity, the activities are ranked in Table 2.3. Highest priority was given to the development of a numerically fast integrator for solving the stiff set of ODEs common to most atmospheric chemistry mechanisms. Considerable progress on this activity has recently been made by Jacobson and Turco (1993) of the University of California at Los Angeles. Their sparse matrix, vectorized Gear solver is being implemented by EPA/AREAL in the UAM-IV and Models-3 system.

Development of a chemical mechanism that addresses the large dynamic range of global trace gas concentrations was the second highest-ranked research activity. This work would support the development of all three classes of CTMs needed in the GTCMS. In a similar fashion, a test of auto-differentiation also addresses all aspects of uncertainty and sensitivity estimation for the GTCMS.

TABLE 2.3. Research Activities in Priority Order, Based on Results of Evaluation by Second Workshop Participants

Rank	Module	Abbreviation	Research Activity
1	Numerics	Fast integrator	Develop numerically fast chemistry integrator
2	Gas Phase	$NO_x/O_3/HC$	Develop appropriate $NO_x/O_3/HC$ chemical mechanism for GTCMS
3	Uncertainty	Autodiff demo	Carry out demonstration of autodifferentiation using test model or module
4	Aqueous Reactions	Aerosols/O_3	Evaluate the effect of aerosols on tropospheric O_3
5	Other Chemistry	OH fields	Evaluate use of short-lived species CTM in providing OH fields for use in long-lived and aerosol CTMs
6	Emissions	Emissions inven	Develop emissions inventories required for GTCMS
7	Aerosols	Evaluate moments	Evaluate species-resolved moments method
8	Aerosols	Moments/species	Develop species-resolved moments of aerosol-size distributions
9	Other Chemistry	Interfaces	Develop interface between short-lived, long-lived, and aerosol CTMs
10	Surface Exchange	Hydro/met synth	Synthesize into consistent framework models for hydrology, surface exchange, emissions, and deposition
11	Aqueous Reactions	Clouds/O_3	Evaluate effects of clouds on tropospheric O_3
12	Transport/Grid System	Irreg grid chem	Develop irregular grid method for chemistry
13	Subgrid Cloud Processes	Subgrid variability	Develop treatment of subgrid variability in clouds and cloud processes
14	Dynamics	Strat/tropo exch	Stratosphere/troposphere exchange
15	Actinic Flux	Update J-values	Update actinic flux J-value look-up tables
16	Scavenging	Aerosols/clouds	Processing of aerosols by clouds using moments method
17	Aerosols	Primary emissions	Emissions of primary particulate and aerosol precursors
18	Gas Phase	Grid res/NO_x	Evaluate effect of grid resolution on NO_x cycling and O_3 production
19	Gas Phase	Chem/aerosol	Develop chemical module for aerosol CTM
20	Clouds	Clouds/CH_4/CO	Role of clouds in CH_4/CO cycle vis-a-viz HCHO and organic acids
21	Nucleation	Mixed aerosols	Parameterize nucleation on mixed aerosols
22	Other Chemistry	DMS chemistry	Evaluate adequacy of gas-phase oxidation mechanisms of DMS and resulting production of CCN
23	Dynamics	Nonlocal	Planetary boundary layer/turbulence nonlocal closure

Rankings below third place change when different scores are used. However, evaluation of the use of the short-lived CTM to generate oxidant fields for the long-lived and aerosol CTMs ranked high by all measures. Work on emissions inventories and methods to estimate emissions also ranked high.

3.0 DECISION ANALYSIS FOR MODEL DESIGN AND RESEARCH PRIORITIZATION

Presented in this section are the underlying rationale that was used to create a vision of the GTCMS, a process for identifying and prioritizing needed research, and a methodology for optimizing the value of a research program, given limited resources. Formal decision analysis methods are described for each of these three activities. The multiattribute utility (MAU) theory is used to identify an optimal vision of the GTCMS and to identify needed research. MAU theory, in combination with value-based planning (VBP), is used to optimize the needed research activities. An optimal GTCMS vision and prioritized set of research activities were generated using MAU in the two workshops. This section describes the decision analysis process and some key results, along with additional activities that would build on these results and make possible the realization of a research and development program.

3.1 OVERVIEW OF THE DECISION PROCESS

Figure 3.1 shows the sequence of decisions used to identify the research needed to develop the GTCMS, along with some of the major considerations that bear on those decisions and their interrelationships. Rectangles represent decisions; ovals represent factors outside direct control. Bolded rectangles represent major decisions. The general flow of the process is along the diagonal from upper left to lower right. The main steps in the process are 1) develop a vision for the GTCMS, 2) determine the science requirements to create the model, 3) define the research needed to produce the model, and 4) prioritize the proposed research.

3.1.1 Vision of Model

The first step in the process is to develop a vision of the model. The vision represents the best collective judgment of the research team as to what the ultimate product of this research effort should be. By clearly specifying the model development goal, it then becomes possible to identify, in a logical manner, what research is needed to attain it. Shown in Figure 3.1 by arrows

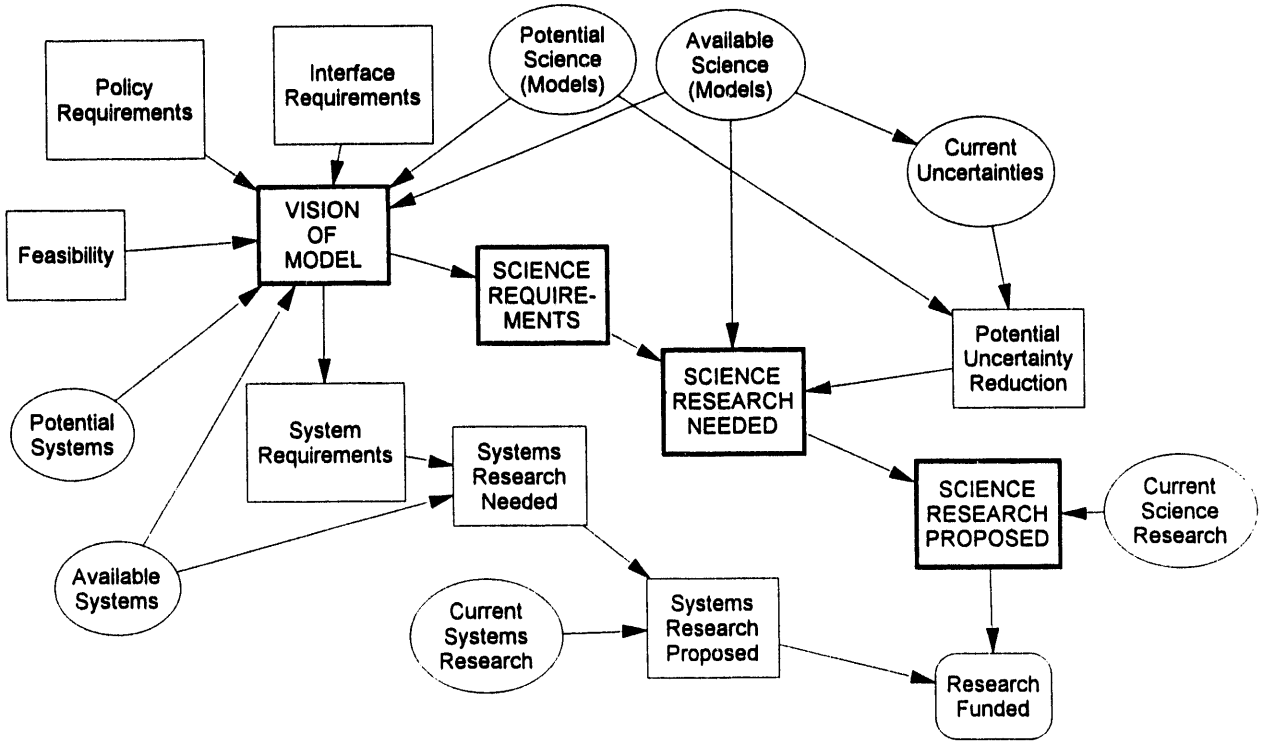

FIGURE 3.1. Logical Flow of Activities to Identify Research Needed to Develop a Global Tropospheric Chemistry Modeling System

pointing to the box labeled "Vision of Model" are the various considerations that enter into the formulation of the vision.

The initial basis for a vision is the science that is available. For each potential module needed to make up the GTCMS, modules may already exist that could be used to perform the required function(s). Collectively, these define what is currently possible and provide a foundation from which the vision evolves.

Potential science represents possible ways to create a GTCMS that improves on the science that is currently available. Potential science can consist of modules that are in various stages of development or alternative approaches for performing a particular module's function. These represent possibilities for expanding the envelope of potential models to the leading edge of what is possible to create within the next few years.

Feasibility addresses the likelihood that the necessary research and development could be carried out within the specified time frame, here defined to be 3 years. Enhancing existing modules and models with potential science may generate some wonderful ideas; however, some of these may need to be tempered with a dose of reality. Some ideas may border on science fiction and others, while sound, may require an extensive effort to carry out, which may not be possible in the allotted time frame. Thus, feasibility considers the risk that may be associated with an ambitious research program that may be required by a given configuration of the model.

Whatever model is chosen, it must be implemented in some hardware environment. The hardware could range from a workstation to a supercomputer; it may be a sequential processor or a parallel processor; less complex models may be better suited to a workstation environment; some models may be better adapted to parallel processors than are others. Consequently, it is necessary to consider what hardware is available and what may be forthcoming.

The GTCMS is envisioned to be a component, albeit a central and essential one, of a more inclusive global change model. Such a model must meet the needs of the policymakers it is being designed to serve. However, it is difficult to predict in advance what the policymakers' concerns will be, and these needs tend to evolve over time. Consequently, this requires that the model be designed to be as flexible as possible in terms of answering a variety of questions under a variety of assumptions and/or background conditions. Every assumption about the model is a potential parameter to be adjusted by the policy user. The ideal model would allow the user to create a variety of scenarios in which to play "what-if" games; realism would be maintained by cautioning against the creation of logically inconsistent conditions. In addition, the ideal model would have the flexibility to interface with the larger global climate change policy modeling system, of which it is one component. This larger system will most likely be identified and/or created after the GTCMS; thus, the GTCMS needs to be adaptable to take a variety of potential input forms and also to provide a variety of outputs and formats as may be required. Also to be considered is the fact that, as a global model, the input data may come from a variety of sources. The ideal

model will be capable of accepting this variety of data either directly or indirectly or via some type of add-on translation capability.

3.1.2 Science Requirements

As shown in Figure 3.1, the next step in the process is to determine the science requirements of the model. A clear specification of the model vision leads directly to the science requirements needed to support its realization. This is the science that makes up the various modules that define the GTCMS. Consideration of the vision for the GTCMS also leads to the identification of system requirements; however, it was decided to focus the design effort on identifying the scientific research necessary to construct a GTCMS.

3.1.3 Science Research Needed

The next step in the sequence shown in Figure 3.1 is to determine the scientific research that is needed for developing and implementing the GTCMS. This depends on what science is required and what is available. Decisions regarding research needed should also consider the potential for reducing uncertainty. The potential for uncertainty reduction depends on the difference between the current uncertainties in the existing science and the anticipated uncertainties in the potential state of science of the options that were selected for each module to create the vision of the model.

3.1.4 Science Research Proposed

Two decisions are necessary to establish what research to propose. In the first decision, areas of research must be selected that are essential to the realization of the model and/or that have the greatest potential for improving currently available science. Decisions regarding which areas need additional research should be based on an evaluation using a well-defined set of criteria that include consideration of research that is currently being carried out elsewhere. In the second decision, proposals must be chosen for specific research projects within each defined research area. Criteria for evaluating proposals should include considerations, such as the prior experience and capabilities of the research organization, in addition to the value of the research. Decision analysis methods can be applied to both decisions. By applying optimization techniques based on decision analysis, one or more

portfolios of research projects can be defined that account for various budget scenarios.

3.2 DECISION ANALYSIS USING MULTIATTRIBUTE UTILITY THEORY

The MAU theory was used to identify an optimal GTCMS vision, to prioritize research needs, and is integral to the recommended process for the optimization of portfolios of research activities. This section provides a description of the main steps in the MAU process, as well as some brief, but general, comments concerning its practical application. Subsequent sections provide additional guidance, as necessary, for the specific applications described.

The MAU decision analysis process consists of the following steps:

- identify objectives to be achieved
- identify alternatives that meet the objectives
- develop measures and values functions for each objective
- develop relative weights for the objectives based on their relative importance
- evaluate alternatives using an objective function and relative weights.

Objectives are statements of what we want to achieve (Keeney 1992). For example, if one were buying a car, some objectives might be to maximize performance, minimize cost, and maximize good looks and styling. For evaluation purposes, objectives are put in a hierarchical form, starting with the overall goal and the fundamental objectives that are further divided into more specific components of those objectives until one arrives at the level of criteria or performance measures. In the example for buying a car, performance might be further divided into acceleration and handling.

The identification of alternatives requires a high level of creativity and should be guided by the overall goal and the specific objectives to be achieved (Keeney 1992). A varied set of alternatives increases the likelihood that a near-optimal alternative will be selected.

Performance measures (also called criteria or attributes) specify the degree to which the objectives are achieved. For example, in the car-purchase scenario, acceleration could be measured by the time (in seconds) required to accelerate from 0 to 60 mi/h. The number of seconds is a natural scale. Often, natural scales are not available, in which case scales must be constructed. Scales should be constructed so as to minimize ambiguity as to what is meant by a given level of performance.

Value functions measure the importance of a criterion. These functions take as their input the various levels of performance and typically map them into the range 0 to 1, or sometimes 0 to 100. The importance of an objective may not be linear with its scale. In the acceleration example, an improvement from 15 to 10 s may be judged more valuable than an improvement from 10 to 5 s. For constructed scales, it is common practice to only specify the composite function of performance level with value. Thus, for example, a score of 75 would specify both the level of performance and the value of that level of performance on a 0 to 100 scale.

Whereas value functions capture the importance of a single criterion, weights capture the tradeoffs among criteria. For example, how much gas mileage is one willing to give up for an additional second-quicker acceleration from 0 to 60 mi/h. Weights logically depend on the ranges in performance over which the alternatives vary. A common mistake is to specify the relative importance of objectives in a "top-down" process without considering the ranges in performance over which the alternatives can vary. A better procedure is to develop weights for criteria in a "bottom-up" process, by taking into account the anticipated performance range for various alternatives over all criteria. "Swing weighting" is a standard decision analysis procedure for accomplishing this. Swing weighting is further described in Section 3.5.

Finally, a mathematical expression, called an objective function, is needed to roll up all the above judgments into a single number by which the alternatives can be compared. The correct functional form for the objective function depends on the preferential independence relationships among the criteria (Keeney and Raiffa 1976).

3.3 APPLICATION OF DECISION ANALYSIS FOR MODEL DESIGN

Selecting a "vision of model" is critical to the creation of a GTCMS. The vision will guide future decisions and determine what research is needed and to what extent. Thus, determining a model vision is a key decision, and a formal decision analysis approach is applied.

An analysis of the functions and requirements of a GTCMS was used to identify its modular components. The selection of particular models for these modular components provides the realization of a GTCMS vision. This section discusses the use of different areas of design emphasis to guide the selection of module options, the criteria for evaluating those selections, and the methods for quantifying the evaluation of alternative designs using the MAU theory.

3.3.1 Chemical Transport Model Alternatives

A formal process was used to identify a variety of model visions that were then evaluated to determine which model is best suited to the needs of policymakers. A shell was provided (see Appendix B) that consisted of the various modules that were identified as being the necessary components of a GTCMS. Alternative approaches were identified that could perform the necessary functions for each of the modules. The selection of an alternative for each module in the shell, when taken collectively, defines a candidate vision for a CTM. Workshop participants also had the opportunity of identifying additional options and/or needed modules.

The design of the CTMs must satisfy a variety of objectives and functional requirements and must consider development constraints as well as the needs of the larger policy analysis system. Many of these objectives are conflicting and require tradeoffs. For example, a simple model is unlikely to maximize its predictive validity. Furthermore, it was not clear in the initial stages of the study which of these requirements was most important. Thus, the strategy chosen was to create a rich variety of potential visions of a CTM that could be evaluated against criteria to be developed. Then, by having experts weight the importance of the criteria and evaluate the potential models against these criteria, the model with the best overall value to

the GTCMS would emerge. The success of this strategy requires a variety of potential models be available for evaluation.

A wide variety of potential CTMs were identified by creating candidate models representing a variety of design emphases. Five design emphases were identified:

- maximize predictive validity and be representative of the best state-of-the-art science
- maximize simplicity while still meeting the performance requirements
- minimize cost of development by using available models
- maximize feasibility by only requiring additional research and development with a minimum of risk
- optimize balance among all the above considerations in the design of the model.

These design emphases are intended to be partially mutually exclusive. The tradeoffs between one or more design emphases were addressed as options for the various modules chosen. Model visions were created for each of these design categories. This resulted in a rich set of visions that could then be formally evaluated. Criteria were developed to choose a vision that would best serve the needs of the policy-making community and be feasible to develop within a 3-year time frame. The evaluation process was carried out within the framework of the MAU theory.

3.3.2 Evaluation Criteria

The criteria developed to evaluate the visions are shown in Table 3.1. Seven criteria were identified as having an impact on the value of a CTM. Most of these scales were constructed from a series of scenarios that represent different levels of performance on the criteria. Each of these scenarios is directly associated with some number in the range of 0 to 100 that represents the value of that level of performance. Thus, for example, a predictive validity score of 70 represents the value to the GTCMS on a scale of 0 to 100 of an R-squared of approximately 0.4 on a scale ranging from 0.1 to 0.7.

For many scales, the level of performance of a particular CTM may be difficult to judge, and one might wonder why define the scale in such a

TABLE 3.1. Criteria Used to Evaluate Potential Chemical Transport Models

Predictive Validity: Measures how well modules simulate the real-world processes that they are designed to model, as measured by the squared correlation coefficient between the predicted and observed values and the absolute value of their difference.

Short-Lived Species: Estimate the average validity, as defined above, across those modules that are most important for predicting the concentration and distribution of short-lived greenhouse gases.

100 High, average R-squared is approximately 0.6 to 0.7, and the absolute value of the difference between observed and predicted values is approximately 1.5 or less.

70 Medium, average R-squared is approximately 0.3 to 0.5, and the absolute value of the difference between observed and predicted values is approximately a factor of 5.

0 Low, average R-squared is approximately 0.1 to 0.2, and the absolute value of the difference between observed and predicted values is approximately a factor of 10.

Long-Lived Species: Estimate the average validity, as defined above, across those modules that are most important for predicting the concentration and distribution of long-lived greenhouse gases.

100 High, average R-squared is approximately 0.6 to 0.7, and the absolute value of the difference between observed and predicted values is approximately 1.5 or less.

70 Medium, average R-squared is approximately 0.3 to 0.5, and the absolute value of the difference between observed and predicted values is approximately a factor of 5.

0 Low, average R-squared is approximately 0.1 to 0.2, and the absolute value of the difference between observed and predicted values is approximately a factor of 10.

Aerosols: Estimate the average validity, as defined above, across those modules that are most important for predicting the distribution and concentration of aerosols.

100 High, average R-squared is approximately 0.6 to 0.7, and the absolute value of the difference between observed and predicted values is approximately 1.5 or less.

70 Medium, average R-squared is approximately 0.3 to 0.5, and the absolute value of the difference between observed and predicted values is approximately a factor of 5.

0 Low, average R-squared is approximately 0.1 to 0.2, and the absolute value of the difference between observed and predicted values is approximately a factor of 10.

Other Modules: Estimate the average validity, as defined above, across all the other modules.

100 High, average R-squared is approximately 0.6 to 0.7, and the absolute value of the difference between observed and predicted values is approximately 1.5 or less.

70 Medium, average R-squared is approximately 0.3 to 0.5, and the absolute value of the difference between observed and predicted values is approximately a factor of 5.

0 Low, average R-squared is approximately 0.1 to 0.2, and the absolute value of the difference between observed and predicted values is approximately a factor of 10.

Maximize Computational Speed: Measures the average complexity of the individual modules, as reflected by the number of computer operations necessary to carry out the calculations factored by degree of parallelism.

100 Fast--measured by few operations per time steps.

50 Moderate--measured by a moderate number of operations per time steps.

0 Slow--measured by many operations per time steps.

TABLE 3.1. (contd)

Maximize Potential for Uncertainty/Sensitivity Analysis: Measures the potential for quantifying uncertainty and the potential ease of sensitivity analysis.

100 The amount of uncertainty in each of the modules can be quantified and there are easy methods available for carrying out sensitivity analysis.

80 Approximately 80% of the modules produce quantitative estimates of uncertainty and there are methods available for carrying out sensitivity analysis.

50 Approximately 50% of the modules produce quantitative estimates of uncertainty and there are methods available for carrying out sensitivity analysis OR approximately 80% of the modules produce quantitative estimates of uncertainty and sensitivity analysis can be carried out only with great difficulty.

0 Less than 20% of the modules produce quantitative estimates of uncertainty and sensitivity analysis can be carried out only with great difficulty.

Closeness to Best Science: Measures how close the model is to the best science.

100 80% of the models represent state-of-the-art science and the remainder is good science.

70 40% of the models represent state-of-the-art science and the remainder is good science.

0 10% of the models represent state-of-the-art science and the remainder is good science.

Flexibility: The extent to which the system can address a variety of policy issues.

100 The system can be easily adapted to answer a large variety of questions.

50 The system can be modified with reasonable effort to answer a standard set of questions.

0 The system is designed to answer a specific set of questions and would be difficult to modify.

Feasibility of Required Research: Measures the likelihood that the research can be successfully carried out within the allotted 3-year time period.

100 Nearly certain that all the required research and development will be successfully accomplished within 3 years.

80 Approximately 80% of the required research is very "straightforward;" remaining 20% is somewhat problematic.

50 50% of the required research is very "straightforward;" 50% is somewhat problematic.

20 Approximately 20% of the required research is very "straightforward;" remaining 80% is somewhat problematic.

0 All the required research is based on highly speculative ideas that may or may not work.

Cost of Research and Development: Measures the cost in person-years for additional research and development to create the model as configured.

100 Entire model can be developed for use as a policy model with approximately 12 person-years of effort. For example, 12 modules require 1 person-year each to develop.

85 Total effort required for development as a policy model is approximately 18 person-years.

70 Total effort required for development as a policy model is approximately 24 person-years.

55 Total effort required for development as a policy model is approximately 36 person-years.

15 Total effort required for development as a policy model is approximately 48 person-years.

0 On average, each of the 18 modules will require 3 person-years to develop, for a total approximate cost of 54 person-years of research.

precise manner. This is done to provide a clear definition of the scale that will pass the "clarity test." This test requires that a clairvoyant, with knowledge of all events, past, present, and future, would be able to unambiguously assign a score for the criterion (assuming that the CTM vision is also clearly defined). Definitions that pass the clarity test serve the function of ensuring that everyone is in agreement with what is meant by a particular criterion. The actual judgment of performance level is based on the best available scientific judgments. These judgments may be based on limited data; but, by clearly defining the concept, consistency of judgment is improved, and so is the overall validity of the process. Additional comments on the criteria/scales (see Table 3.1) follow:

- Predictive Validity--In cases where a model or module did not yet exist, predictive validity was based on expected performance.
- Computational Speed--Computational speed was designed to be independent of the hardware system in which the model was realized.
- Potential for Uncertainty/Sensitivity Analysis--While this is difficult to estimate, it is important to policy analysts and was included in the evaluation.
- Closeness to Best Science--It is possible for a model to have good predictive validity without actually providing a realistic model of the processes that it is designed to predict. Also, the closer the model is to state-of-the-art science the more likely it is to be acceptable to the science community. Thus, this criterion captures important considerations besides those of predictive validity.
- Flexibility--This was necessary to ensure the ability of the model to address a variety of policymaker concerns, many of which are not currently known.
- Feasibility of Required Research--This provides assurance that the model can be created within the allotted time frame.
- Cost--Cost estimates were based on CTM visions defined by a collection of modules. Some of these modules already exist, others will need research to create. The cost estimates were based on the discrepancy between existing science and what would be needed to create a particular model.

3.3.3 Option Generation

Multiple visions for the three classes of CTMs, based on the different design emphases, were created in the second workshop. The process consisted

of independent judgments regarding option selection, followed by discussion within small groups dedicated to a particular class of CTM. These selections were then compared in small working groups to arrive at a consensus for each of the design emphases. This resulted in a rich set of models that could then be formally evaluated to choose a vision that would best serve the needs of the GTCMS and be practical to develop within the time constraints.

Tables 3.2 through 3.4 present, for each of the three classes of CTMs, the options selected for each of the design emphases. Initial and final optimal designs tended to be very similar. The final optimal design was generated to provide an opportunity for revision after considering the specific design emphases. Designs emphasizing predictive validity tended to include those options with the greatest process detail. In contrast, designs that emphasized computational speed used options with greater degrees of process parameterization. Designs highlighting low cost of development and feasibility usually made use of existing options and parameterized processes.

3.3.4 Option Evaluation

Participants at the second workshop evaluated the various options for each class of CTM. This was carried out in three small groups--each group being dedicated to a particular class of CTM. Group members represented a variety of backgrounds, thus ensuring that the relevant expertise was available. The results of these evaluations are presented in Table 3.5. The design emphases for the three classes of CTMs are shown along the top; the rating criteria are shown to the left. The body of the upper portion of the table gives the score each version received when rated by the participants on the criteria. The scores were averaged across the participants in the relevant small group. The scores, while averages, are a result of discussions leading to group consensus in most cases. The weights represent value judgments concerning the relative importance of the various criteria. The weights were assigned using the swing-weighting process described in Section 3.5. The weights used in the analysis are the average of the weights assigned by all the workshop participants, and therefore, reflect their collective judgment.

The value associated with each version was calculated by multiplying the score by the weight and summing over all the criteria, except feasibility.

TABLE 3.2. Options Selected for Alternative Visions: Short-Lived Species Chemical Transport Model

	Short-Lived Species Model			
	Optimal	Validity	Speed	Cost
Chemistry				
Aqueous Rxns	S+radical rxns	S+radical rxns	None	None
	NOy/O3/H202	NOy/O3/H202		
Aqueous Size	Bulk	Size resolved	None	None
Gas-Phase Rxns	Multiple lumped HC	Explict HC	Background	Background
	S/NOx/O3	S/NOx/O3/Cl	NOx/O3	NOx/O3
Aerosols	Bulk/Internal mixing	Size & spec res	None	None
Subgrid	Plume-grid	Plume-statistical	Instant	Instant
Cloud Processes				
Microphysics	Bulk	Size resolved	None	None
Scavenging	Bulk	Size resolved	None	None
Activation	Bulk	Size resolved	None	None
Convective Clouds	Coupled/conv adj	Coupled/3rd G	None	None
Subgrid	Fraction	Coupled/GCM	None	None
Dynamics				
PBL/Turbulence	TKE 1.5	2nd O Kzz	1st O Kzz	1st O Kzz
Strato/Tropo Exch.	Dyn w/o chem	Dyn w/ chem	B.C.	B.C.
Actinic Flux				
Radiative Transfer	On-line clds	On-line calc	Look up	Look up
Subgrid	Fraction	3-D effects	Uniform	Uniform
Surface Exchange	Sfc exchange	Sfc exchange	Constant	Constant
Numerics				
Transport	1D FEM	2D FEM	1D FEM	1D FEM
Chemistry	QSSA	New solver	QSSA	QSSA
Grid	Irregular	Adaptive	Uniform	Uniform

TABLE 3.3. Options Selected for Alternative Visions: Long-Lived Species Chemical Transport Model

	Long-Lived Species Model					
	Optimal	Validity	Uncertainty	Speed**	Cost	Feasibility
Chemistry						
Aqueous Rxns	None	None	None	None	None	None
Aqueous Size	None	None	None	None	None	None
Gas-Phase Rxns	CO, CH4	CO, CH4	CO, CH4	CO, CH4	CO, CH4	CO, CH4
OH	Prescribed	Seasonal	Prescribed	Prescribed	Prescribed	Prescribed
Aerosols	None*	None*	None	None	None	None
Subgrid	Instant	Instant	Instant	Instant	Instant	Instant
Cloud Processes						
Microphysics	None	None	None	None	None	None
Scavenging	None	None	None	None	None	None
Drop Activation	None	None	None	None	None	None
Convective Clouds	Cpld GCM†	Cpld GCM†	Cpld GCM†	None##	None	None
Subgrid	Cpld GCM†	Cpld GCM†	Cpld GCM†	None##	None	None
Dynamics						
PBL/Turbulence	1st O Kzz	Online w/GCM	1st O Kzz	1st O Kzz	1st O Kzz	1st O Kzz
Strato/Tropo Exch.	Research	GCM†	Max uncert	GCM†	B. Cond.	B. Cond.
Actinic Flux						
Radiative Transfer	Look up	GCM†	Look up	Look up	Look up	Look up
Subgrid	Look up#	GCM†	Look up	Look up	Look up	Look up
Surface Exchange	Emissions	Emissions	Emissions	Emissions	Emissions	Emissions
Numerics						
Transport@	Not Lagran	Not Lagran	Any	Not Lagran	Not Lagran	Not Lagran
Chemistry	Non-stiff	Non-stiff	Non-stiff	Non-stiff	Non-stiff	Non-stiff
Grid	Uniform	Uniform	Uniform	Uniform~	Uniform~	Uniform~

* If no feedback processes

\# Presumes OH distribution is specified

† Use same parameterization as in GCM

@ Moments not needed

** As much computation done off-line as possible

Use GCM wind fields to reflect convection

~ Explore how to degrade horizontal resolution

TABLE 3.4. Options Selected for Alternative Visions: Aerosol Chemical Transport Model

	Aerosol Model					
	1st Optimal	Validity	Speed	Cost	Feasibility	2nd Optimal
Chemistry						
Aqueous Rxns	Sulfur	Sulfur	Sulfur	Sulfur	Sulfur	Sulfur
Aqueous Size	Bulk	Moments	Bulk	Bulk	Bulk	Bulk
Gas-Phase Rxns	Hybrid	Hybrid	OH fields	OH fields	Hybrid	OH fields
	Nox/S	Nox/S			Nox/S	
Aerosols	Moments/ext	Moments/ext	Bulk/ext	Bulk/ext	Moments/ext	Moments/ext
Subgrid	Instant	Instant	Instant	Instant	Instant	Instant
Cloud Processes						
Microphysics	Moments/ice	Moments/ice	Moments/ice	Bulk	Moments/ice	Moments/ice
Scavenging	Moments	Moments	Moments	Bulk	Moments	Moments
Drop Activation	Moments	Moments	Bulk	Bulk	Moments	Moments
Convective Clouds	A&S#	3rd gen	Kuo/1 plume*	W&T@	A&S	A&S
Subgrid	Statistical	Statistical	Fract. area	Fract. area	Fract. area	Statistical
Dynamics						
PBL/Turbulence	TKE 1.5	TKE 1.5	1st order	1st order	1st order	TKE 1.5
Strato/Tropo Exch.	B.C.	B.C.	B.C.	B.C.	B.C.	B.C.
Actinic Flux						
Radiative Transfer	On-line cloud	On-line cloud	None	None	On-line cloud	None
Subgrid	Cld. fraction	Cld. fraction	None	None	Cld. fraction	None
Surface Exchange	Resistance	Resistance	Resistance	Resistance	Resistance	Resistance
Numerics						
Transport	1D FDM/FEM	1D FDM/FEM	1D FDM/FEM	1D FDM/FEM	1D FDM/FEM	1D FDM/FEM
Chemistry	Sillman	Sillman/Potra	Simple†	Simple†	Sillman	Simple†
Grid	Irregular	Irregular	Irregular	Uniform	Irregular	Irregular

Arakawa and Schubert (1974) * Kuo or 1-plume A&S @ Walcek and Taylor (1986) † non-stiff

TABLE 3.5. Ratings by Workshop Participants of the Alternative Chemical Transport Model Visions

CRITERIA		MODELS												
	WEIGHT	LONG LIVED				SHORT LIVED			AEROSOL					
PREDICTIVE VALIDITY		Opt	PV	Unc	SCF	PV	SCF	Opt	Opt	PV	Speed	Cost	Feas	F Opt
Aerosols	0.07	0	0	0	0	70	0	35	78	85	51	45	69	78
Short-Lived Species	0.08	0	0	0	0	80	50	70	58	70	38	35	54	35
Long-Lived Species	0.13	70	70	70	70	80	70	80	0	0	10	0	0	0
Other Modules	0.04	0	0	0	0	0	0	0	0	0	0	0	0	0
TOTAL PV weight	0.32													
COMPUTATIONAL SPEED	0.13	90	80	80	100	0.1	30	10	58	64	86	90	70	78
UNCERTAINTY ANALYSIS POTENTIAL	0.14	60	30	80	30	50	50	50	55	64	76	79	65	73
FLEXIBILITY FOR ALTERNATIVE POLICY USES	0.17	25	25	25	25	75	40	70	51	72	40	40	50	42
CLOSENESS TO BEST SCIENCE	0.23	50	50	50	50	100	5	70	89	93	60	55	72	68
FEASIBILITY		100	100	100	100	10	100	70	80	60	90	95	90	90
VALUE (Σ(WEIGHT*SCORE))														
VALUE 1: PV weights on all modules		45	39	46	42	64	32	55	54	62	50	48	52	51
VALUE 2: 0.32 weight on main module		58	53	60	55	68	35	59	69	78	59	57	65	68
EXPECTED VALUE (VALUE*FEASIBILITY/100)														
E-VALUE 1: PV weights on all modules		45	39	46	42	6	32	38	44	37	45	46	47	46
E-VALUE 2: 0.32 weight on main module		58	53	60	55	7	35	41	55	47	53	54	59	61
EFFORT AND COST (Person-Years)		6	7	8	5	54	12	32	18	26	13	12	16	15

Opt Initial optimal design
PV Emphasis on predictive validity
Unc Emphasis on uncertainty analysis
Speed Emphasis on computational speed
Cost Emphasis on low cost of development
Feas Emphasis on high feasibility of research and development
SCF Designs for speed, cost, and feasibility were identical
F Opt Final optimal design

Value 1 is calculated using each of the four categories of predictive validity treated as a separate criterion. Because of the low scores given to the predictive validity for all but the modules relevant to the particular class of model, the Value 1 calculation deemphasizes the overall importance of the predictive validity criterion.

Value 2 gives more weight to predictive validity by associating the total weight for the predictive validity criterion, with the score given to the group of modules corresponding to the class of model being evaluated. Thus, Value 2 does not favor those designs that could have predictive validity for more than one class of model. The expected value rows show the overall value discounted by the feasibility of carrying out the necessary research and development within the allotted 3-year time frame. This row is the value score multiplied by the feasibility score. Because feasibility is a probability measure, this product is the expected value of each of the CTM visions, where expected value has the standard interpretation used in statistics and decision analysis. The last row of Table 3.5 shows the estimated level of effort required to produce the model in person-years.

Based on the ratings presented in Table 3.5, one vision for each class of CTM was selected as the target for the GTCMS design. For the long-lived species CTM, the initial optimal version and the version for ease of uncertainty analysis score closely, with the latter estimated to require slightly more effort to develop; the optimal design was selected. The group that developed the selections for the short-lived species CTM found many of the design emphases to lead to essentially the same option selections. In the rating of their visions, the final optimal design ranked highest and was selected. Although the version designed to maximize its predictive validity scored very high in all criteria but feasibility, this design was given little chance of being developed within the 3-year time frame and its cost would be high. Rating of the alternative visions for the aerosol CTM resulted in the most feasible and final optimal designs to have very similar ratings and require a modest level of effort; the final optimal design was selected. Option selections for the top-rated alternative visions for each class of CTM in the GTCMS are given in Table 3.6. Collectively, these define the GTCMS and provide the basis for identifying research necessary for its realization.

TABLE 3.6. Options Selected for Top-Rated Alternative Visions

Module	Class of CTM		
	LONG-LIVED	SHORT-LIVED	AEROSOL
	Optimal	Optimal	Optimal
Chemistry			
Aqueous Rxns	None	S+radical rxns+NOy/O3/H202	Sulfur
Aqueous Size	None	Bulk	Bulk
Gas-Phase Rxns	CO, CH4, OH prescribed	S/NOx/O3/lumped HCs	OH fields prescribed
Aerosols	None*	Bulk/internally mixed	Moments/species resolved
Subgrid	Instant dispersion	Plume-in-grid	Instant dispersion
Cloud Processes			
Microphysics	None	Bulk	Moments/ice
Scavenging	None	Bulk	Moments
Drop Activation	None	Bulk	Moments
Convective Clouds	Coupled to GCM	Coupled/convective adjustment	Coupled/Arakawa & Schubert
Subgrid	Coupled to GCM	Fraction	Statistical
Dynamics			
PBL/Turbulence	K-theory, 1st O Kzz	K-theory, 1.5 O Kzz (TKE)	K-theory, 1.5 O Kzz (TKE)
Strato/Tropo Exch.	Research	Dynamic w/o chemistry	Boundary condition
Actinic Flux			
Radiative Transfer	Look up	On-line clouds	None
Subgrid	Look up, OH prescribed	Fraction	None
Surface Exchange	On-line GCM for CO2	Surface exchange model	Resistance
Numerics			
Transport	Not Lagrangian	1D FEM	1D FDM/FEM
Chemistry	Gear	QSSA	Simple (non-stiff)
Grid	Uniform	Irregular	Irregular

3.4 APPLICATION OF DECISION ANALYSIS TO RESEARCH PRIORITIZATION

This section describes the process by which the MAU theory was applied in the second workshop to prioritize the list of research needs (see Table 2.2). The criteria for the prioritization of research activities were designed to maximize the value of research carried out for the creation of the three model visions that were earlier identified as optimal. The criteria and scales are shown in Table 3.7.

Again, the numerical values shown in Table 3.7 combine the performance measures and the value functions. Thus, for Improve Predictive Validity, an increase in the value of R-squared by 0.2 is scored a value of 50 because it

TABLE 3.7. Criteria Used to Evaluate Research Needs

Improve Predictive Validity: Measures the expected improvement in predictive validity.

- 100 R-squared for a module for which RITS are sensitive could increase by approximately 0.5 and/or the difference between the observed and predicted values could greatly diminish.
- 50 R-squared for a module that plays a central role in the overall predictive validity could increase by approximately 0.2 and/or the difference between the observed and predicted could become smaller OR R-squared for a module that plays a peripheral role in the overall validity could increase by 0.5 and/or the difference between the observed and predicted values could greatly diminish.
- 25 R-squared for a module that plays a peripheral role in the overall validity could increase by 0.2 and/or the difference between the observed and predicted values could become smaller.
- 0 There is no potential to improve the science in a module.

Reduce Computation Time: Measures the expected reduction in computation time.

- 100 Time for computationally expensive module could be reduced to 10% of current value.
- 50 Time for computationally expensive module could be reduced to 50% of its current value OR time for a moderately costly module could be reduced by 90%.
- 20 Time for computationally expensive module could be reduced to 80% of its current value OR time for a moderately costly module could be reduced by 50%.
- 0 No potential to reduce computational time.

Scope of Application: Measures the number and importance of the modules for which the research applies.

- 100 Fully supports a module that is very important for several models OR several modules that are very important for a single model.
- 50 Fully supports a module that is somewhat important for several models OR several modules that are somewhat important for a single model.
- 25 Fully supports a single module that applies to one model OR partially supports a module that is very important for several models.
- 0 Only partly supports a single module that applies to one model.

Intrinsic Scientific Value: Measures the value of the research to the scientific community.

- 100 Research has substantial intrinsic scientific value with broad implications beyond the scope of the GTCMS effort.
- 50 Research has intrinsic scientific value with some implications beyond the scope of the GTCMS effort.
- 0 Research has primarily value for the GTCMS effort but little additional scientific value.

Extent of Independent Funding: Measures the extent of funding by EPA would duplicate efforts elsewhere.

- 100 No independent research is currently being funded or there is an opportunity to participate in an ongoing research project.
- 50 Moderate amount of independent research is being funded.
- 0 Research is already fully funded.

Likelihood of Success: Measures probability of success in allotted time.

Estimate probability from 0 to 1.

Cost: Measures person-years, or their equivalent cost in equipment, for completing research and development necessary to incorporate results into model.

Estimate number of person-years.

is thought to have one half of the value of an increase by 0.5, which is scored 100.

Workshop participants individually evaluated the research activities using the criteria and scoring outlined in Table 3.7. Table 3.8 gives the scores averaged over all participants submitting scores for the research activities for each of the criteria. The relative weights for the criteria were determined during the workshop using the swing-weighting technique described in Section 3.5. These are shown across the top row.

Table 3.9 gives the weighted scores and rankings for the 23 research activities. The raw score is based on the weights for the first four criteria. The adjusted scores multiplicatively factor in the scores given for independent funding (Fndg) and likelihood of success (Feas) separately and combined. Except for the two highest-ranked activities, developing a fast integrator for stiff differential equations and developing a chemical mechanism for global-scale photochemistry, either or both of these multiplicative criteria significantly affected the ranking of research activities. For instance, evaluation of the effects of aerosols on tropospheric O_3 ranked 13th in raw score, but because little research in this area is being currently funded, its adjusted score put it at 4th. Alternatively, research into the exchange of mass between the stratosphere and troposphere, which ranked 5th in raw score, dropped to 14th in the combined adjusted score because of a low score on likelihood of success within a 3-year time frame.

The MAU process used here to arrive at the raw and adjusted scores shown in Tables 3.8 and 3.9 enables a ranking of research activities without consideration of the resources required to accomplish the work. Introducing this factor into the decision process requires additional tools. A methodology for prioritizing research and optimizing the utilization of funding resources is described in the next section.

TABLE 3.8. Average Scores for Workshop Participants that Rated Research Activities

	Weight	40	20	18	22			
Module		PRED.	COMP.		SCIENT.	INDEP.	SUCC.	EFFORT
	Research Activity	VALUE	SPEED	SCOPE	VALUE	FNDG	PROB.	(PYrs)
Clouds, Aerosols, Radiation								
Nucleation								
	Mixed Aerosols	44.6	8.2	40.8	61.9	54.6	66.3	3.5
Subgrid Cloud Processes								
	Subgrid Var.	57.7	18.9	56.9	64.2	61.3	69.6	4.6
Aerosols								
	Moment/Species	61.9	20.1	58.5	68.1	59.2	77.3	7.8
	Evaluate Moments	59.6	12.2	52.5	63.3	64.2	78.8	3.7
	Primary Emissions	75.4	8.9	71.5	58.1	46.7	77.1	4.5
Scavenging								
	Aerosols/Clouds	54.2	18.5	48.1	59.2	60.8	73.5	2.7
Actinic Flux								
	Update J-Values	61.7	13.4	67.5	60.8	49.0	82.7	2.4
Chemistry								
Gas Phase								
	NOx/O3/HC	75.9	30.9	77.3	70.5	61.5	82.0	6.5
	Grid Res/NOx	63.2	40.5	63.6	58.2	48.6	70.6	3.3
	Chem/Aerosol	62.5	22.7	50.5	65.5	49.1	71.4	2.3
Emissions								
	Emissions Inventory	76.8	14.5	80.5	56.8	54.5	81.5	2.5
Aqueous Reactions								
	Aerosols/O3	70.5	14.2	50.0	64.1	67.0	75.0	2.9
	Clouds/O3	71.8	13.7	51.8	65.5	59.0	73.5	2.9
Clouds								
	Clouds/CH4/CO	49.5	20.5	44.1	54.1	59.0	70.0	2.6
Chemistry								
	Interfaces	61.4	27.3	65.0	39.5	62.2	78.0	2.6
	OH Fields	73.6	31.4	67.3	59.5	57.5	77.5	3.6
	DMS Chemistry	53.9	12.2	50.0	58.3	46.3	68.1	2.1
Meteorology & Numerics								
Dynamics								
	Strat/Tropo Exch	75.4	13.0	65.8	68.3	56.4	64.1	5.2
	Non-Local	44.5	20.6	50.0	47.5	43.9	73.3	3.1
Surface Exchange								
	Hydro/Met Synth	74.0	11.0	77.0	59.0	52.5	77.8	4.8
Transport/Grid System								
	Irreg Grid Chem	56.7	47.9	71.3	60.0	54.5	73.6	3.5
Numerics								
	Fast Integrator	61.2	86.2	89.6	75.8	70.4	79.2	4.0
Uncertainty								
	Autodiff demo	41.1	48.9	81.5	79.0	65.0	77.5	1.9

TABLE 3.9. Weighted Scores and Rank for Research Activities, Based on Scoring by Workshop Participants

		Weighted Scores				Rank			
Module	Research Activity	Raw Score	Fndg Adj	Feas Adj	Fndg & Feas Adj	Raw Score	Fndg Adj	Feas Adj	Fndg & Feas Adj
Clouds, Aerosols, Radiation									
Nucleation									
	Mixed Aerosols	40	22	27	15	23	21	23	21
Subgrid Cloud Processes									
	Subgrid Var.	51	31	36	22	16	12	18	13
Aerosols									
	Moment/Species	54	32	42	25	12	9	10	8
	Evaluate Moments	50	32	39	25	18	10	15	7
	Primary Emissions	58	27	44	21	10	17	7	17
Scavenging									
	Aerosols/Clouds	47	29	35	21	19	15	19	16
Actinic Flux									
	Update J-Values	53	26	44	21	15	19	8	15
Chemistry									
Gas Phase									
	NOx/O3/HC	66	41	54	33	2	2	2	2
	Grid Res/NOx	58	28	41	20	9	16	11	18
	Chem/Aerosol	53	26	38	19	14	18	17	19
Emissions									
	Emissions Inventory	61	33	49	27	4	7	3	6
Aqueous Reactions									
	Aerosols/O3	54	36	41	27	13	4	12	4
	Clouds/O3	55	33	41	24	11	8	13	11
Clouds									
	Clouds/CH4/CO	44	26	31	18	21	20	21	20
Chemistry									
	Interfaces	50	31	39	24	17	13	14	9
	OH Fields	61	35	47	27	3	5	4	5
	DMS Chemistry	46	21	31	14	20	22	20	22
Meteorology & Numerics									
Dynamics									
	Strat/Tropo Exch	60	34	38	22	5	6	16	14
	Non-Local	41	18	30	13	22	23	22	23
Surface Exchange									
	Hydro/Met Synth	59	31	46	24	6	14	5	10
Transport/Grid System									
	Irreg Grid Chem	58	32	43	23	7	11	9	12
Numerics									
	Fast Integrator	74	52	59	42	1	1	1	1
Uncertainty									
	Autodiff Demo	58	38	45	29	8	3	6	3

3.5 APPLICATION OF DECISION ANALYSIS TO RESEARCH PORTFOLIO SELECTION

This section describes a decision methodology for prioritizing the funding of research areas and activities. By way of example, the method is applied to research activities that were identified in the first and second workshops for the creation of the GTCMS. The methodology, as described, is generally applicable to complex research and development situations. The methodology has its basis in MAU, which has been described earlier, and value-based planning (VBP), which will be described and illustrated here. MAU is used to identify the potential value of proposed research, and VBP is used to optimize portfolios of research according to benefit-cost ratios. The result is a prioritized ranking of proposed research activities for different budget levels. The process is carried out in two stages: 1) to prioritize individual proposals and 2) to prioritize bundles of proposals grouped by topic area. The steps to the process are as follows:

- determine categories of research and generate proposals
- prioritize individual proposals
- generate research program options for each category of research
- prioritize research programs
- determine optimum research portfolios.

3.5.1 Determining Research Needs

The first step in research prioritization is to define what research to request. The exercises carried out in the first and second workshops provided illustrative examples of specific areas in which research is needed for developing the GTCMS. It is recommended that the requested research cover several major research categories. These could be the general areas of research identified in the workshops or other categories as deemed appropriate. For purposes of discussion, in this section we will assume that requests for proposals (RFPs) are written for the following major research areas:

- clouds, aerosols, and radiation
- chemistry

- meteorology
- numerics.

It may prove advantageous to develop RFPs that address subareas within these broad categories. Thus, in this example, for the category clouds, aerosols, and radiation, the following subcategories were identified in the workshop:

- activation
- subgrid cloud processes
- aerosols
- scavenging
- actinic flux.

The illustrative examples of research activities, specific to each of these areas, that were developed in the second workshop are presented in Appendix C.

The next step in the prioritization process is to generate proposals through the release of an RFP. Proposals must contain all the information necessary for their evaluation; consequently, the RFPs need to specify that proposals contain the specific information that they will be evaluated on. While it may not be desirable to list specific criteria and scales in the RFP, it probably is desirable to list, in general terms, the basis for the evaluation and, possibly, which areas will be given the most consideration or weight. Consequently, thought should be given to the proposal evaluation criteria at the time the RFPs are generated. These evaluation criteria will be subject to the regulations and practices specific to the funding organization. It is recommended that the evaluation criteria include technical criteria that address the usefulness of the research in creation of a GTCMS, criteria that address the qualifications of the research institution and the principal investigators, and cost information. Criteria will be discussed further in the next section.

3.5.2 Prioritizing Individual Proposals

The evaluation criteria usually identify what is important in the evaluation of the proposals. Individual proposals in most institutions are evaluated on the technical merit of the research, the ability of the research

institution to successfully support the research, the ability of the scientists to carry out the research, and the cost to the funding organization. Criteria for evaluating the technical merit of research specific to the creation of a GTCMS were developed for the exercise carried out in the second workshop. The technical criteria and their scales were shown in Table 3.7. The scales were constructed to clearly define specific levels of performance for each of these criteria. It is important that all criteria have scales that unambiguously define their levels of performance and that are broad enough to include the best- and worst-possible scenarios. Also shown in Table 3.7 are the numbers ranging from 0 to 100. These numbers represent the value of the associated performance level for the overall goal of supporting research necessary for the development of the GTCMS. Thus, value functions have been incorporated directly into the scales so that the numbers not only represent the level of performance but also the value associated with achieving that level of performance.

The technical criteria and scales developed in the second workshop may be used directly as shown or they may be modified as required for the specific application. In any case, it will be necessary to develop additional criteria and scales to evaluate the ability and qualifications of the research institution and the principal investigators. Examples of some possible additional criteria are the following:

- scientific merit of proposal, including logic of research plan
- qualification and support of research institution
- academic qualifications of key personnel
- time commitment of personnel
- project management capability and organization.

To capture the relative importance of the complete set of evaluation criteria, weights must be established. Swing weighting is a standard decision analysis process that is recommended for establishing the weights. This method results in the criteria weights reflecting the ranges over which the alternatives can vary and correctly ties the weights to the scales. The

elicited swing weights reflect the tradeoffs among the criteria in their respective units.

The swing-weighting procedure is a bottom-up process, in which the criteria within a particular category, or subcategory, are first ranked and then their ratios of relative importance determined. The swing process requires that the worst- and best-performance levels for each of the criteria be identified. The first step in the swing-weighting process is to consider a situation in which a hypothetical proposal would score at the worst level for all criteria within a particular category. Then imagine that if the hypothetical proposal could be improved to the best level on one of the criteria, for which criterion would this represent the most valuable improvement, the second most valuable, etc. This provides the basis for the rankings. Ratio judgments of the relative importance of the criteria are then obtained. The process is repeated for each of the categories and then extended to obtain judgments of relative importance across categories. The ratios of relative importance ascribed to the various criteria should then be normalized, so that the weights across all the criteria sum to 1. A spreadsheet, developed prior to obtaining the necessary judgments will greatly facilitate the process. The first column in the spreadshee d list the criteria; the second and third columns should show the wors est levels of performance for each criterion; next should be columns for ranking the criteria and rating them with the ratio judgments of relative importance. The spreadsheet can easily be developed to compute normalized weights from the raw ratio judgments of relative importance.

Weights should be obtained from each member of the evaluation panel and then averaged, providing that the differences between panel members are not too great. Substantial differences in assigned weight should be discussed and resolved.

Specific evaluation assignments will depend on the breadth of knowledge of the individual evaluators and the number of proposals received. If the evaluators are competent to rate proposals from all of the major research categories and the number of proposals is not too great, then all evaluators can rate all proposals on all criteria. The scoring should initially be made independently and then be compared. If there are large differences in

scoring, these should be discussed, with the goal to reach consensus. The results from the individual evaluators can then be averaged to determine the ratings or score. If the evaluators do not feel competent to rate proposals from all areas, and/or the number of proposals is too large, then individuals or small teams can be assigned to rate proposals in specific research categories. It will be especially important in this case that the scales for evaluating each criterion be clearly defined. Only then will meaningful comparisons of proposals across research categories be possible. Clearly defined scales should always be developed, so that proposals can be given objective ratings rather than relative comparisons, which become very difficult when there are many proposals to rate.

Each proposal is then scored on each of the criteria according to defined scales. These scores should then be combined with the weights to determine the overall rating of the proposal. Generally, the weight and score are multiplied for each criterion and the product summed over all criteria to determine the overall rating for each individual proposal. However, this assumes the criteria are preferentially independent. In some cases, the overall rating is better reflected in a model in which some of the criteria are multiplied (Keeney and Raiffa 1976). In any case, the process is greatly facilitated by the creation of a worksheet on an electronic spreadsheet. Table 3.10 shows the layout of a typical worksheet for scoring individual proposals. Fictitious data are used to illustrate the process. The worksheet identifies the evaluator (AB) at the top, with the list of the proposals being evaluated in the first column. Subsequent columns contain the scores assigned by the evaluator to each criterion for each proposal. Table 3.11 shows a spreadsheet that calculates the overall rating of an individual proposal, using the scores from several evaluators. This spreadsheet lists the criteria in the first column, then the weights, a column for the scores from each of the evaluators, and a column for the average of the evaluators' scores. The last entry in the last column is the calculated overall score, based on the average scores and the weights. In this example, the average score is multiplied by the weight for the first eight criteria; these products are then summed. This sum is then multiplied by the independent funding score divided by 100, and the resulting product is then multiplied by probability of

TABLE 3.10. Typical Worksheet for Scoring Proposals

			SCORING SHEET FOR INDIVIDUAL PROPOSALS								
Evaluator:AB	CATEGORY: CLOUDS, AEROSOLS, RADIATION										
		PRED.	COMP.		SCIENT.	INDEP.	SUCC	ACAD	INSTIT	PLAN	TIME
PROPOSAL:		VALUE	SPEED	SCOPE	VALUE	FNDG	PROB.	QUAL	SUPPORT	LOGIC	COMMIT
Activitation											
Mixed Aerosols		25	20	75	50	75	80	90	80	85	100
Subgrid Cloud Processes											
Subgrid Var. L1		50	20	50	75	75	90	70	90	60	90
Subgrid Var. L2		75	50	50	75	90	90	50	20	95	100
Aerosols											
Moment/Species L1		50	75	75	90	90	90	40	90	80	50
Moment/Species L2		50	90	80	100	100	75	65	40	95	40
Evaluate Moments		75	0	50	90	90	75	25	100	70	100
Primary Emissions		20	0	50	50	50	90	85	55	90	70
Scavenging											
Aerosols/Clouds		50	50	50	50	90	75	50	40	90	60
Actinic Flux											
Update J-Values		50	75	50	50	75	90	85	35	95	70

TABLE 3.11. Spreadsheet to Determine Overall Value of Individual Projects

CATEGORY: Clouds, Aerosols, Radiation					
Proposal: Mixed Aerosols				**Cost: 350K**	
			Evaluator		
Criteria:	**Weight**	A B	C D	E F	Average
Predictive Value	0.24	80	65	50	65
Computational Speed	0.12	20	25	35	27
Scope	0.10	90	70	75	78
Scientific Value	0.14	85	60	100	82
Academic Qualifications	0.15	90	70	90	83
Institutional Support	0.05	80	75	85	80
Research Plan Logic	0.10	85	75	90	83
Time Commitment	0.10	100	90	100	97
Independent Funding	na	100	80	70	83
Probability of Success	na	0.9	0.7	0.6	.73
				Value:	**44**

success. In this example, the overall value is best served by a model in which the last two criteria act as discounting factors for the value determined by the other criteria.

Table 3.12 is illustrative of a spreadsheet that compares the individual proposals in a given research category. A proposal comparison spreadsheet should be created for each of the major research categories. So, for this example, there would be four spreadsheets. The spreadsheet shows the overall value of the proposals, the proposed cost, and the value to cost ratio (V/C). The lower portion of the spreadsheet gives an initial ranking of all the proposals within a given category based on the value-to-cost ratio. This portion of the table was created by duplicating the top portion and sorting the rows based on the value-to-cost ratio, with the highest ratio in the top row. An additional column added to this portion of the spreadsheet gives the cumulative cost of funding any number of proposals based on the value-to-cost rankings. (In practice, all of these spreadsheets can be created on a single large spreadsheet or the small individual spreadsheets can be "linked.")

TABLE 3.12. Comparison of Individual Research Proposals

CATEGORY: Clouds, Aerosols, Radiation					
PROPOSAL:					
		VALUE	COST(K)	V/C	
Activitation					
	Mixed Aerosols	44	350	0.13	
Subgrid Cloud Processes					
	Subgrid Var. L1	55	200	0.28	
	Subgrid Var. L2	65	460	0.14	
Aerosols					
	Moment/Species L1	41	400	0.10	
	Moment/Species L2	54	780	0.07	
	Evaluate Mmoments	28	370	0.08	
	Primary Emissions	21	450	0.05	
Scavenging					
	Aerosols/Clouds	70	270	0.26	
Actinic Flux					
	Update J Values	35	240	0.15	
PROPOSAL:					CUM
		VALUE	COST(K)	V/C	COST
	Subgrid Var. L1	55	200	0.28	200
	Aerosols/Clouds	70	270	0.26	470
	Update J Values	35	240	0.15	510
	Subgrid Var. L2	65	460	0.14	700
	Mixed Aerosols	44	350	0.13	810
	Moment/Species L1	41	400	0.10	750
	Evaluate Moments	28	370	0.08	770
	Moment/Species L2	54	780	0.07	1150
	Primary Emissions	21	450	0.05	1230

3.5.3 Creating Research Programs

Ultimately, decisions on research are made from a collection of research proposals whose overall value is not necessarily a simple sum of the values of the individual projects. For example, two proposals with high value-to-cost scores may have large overlaps in the proposed research, so that their values would not be additive. Consequently, what is needed is that the evaluators

generate recommended combinations of proposals that make sense for each of the major research categories. Furthermore, research needs to be optimized for different levels of budgetary constraints; consequently, each group of evaluators needs to recommend combinations of proposals for several different funding levels. Typical funding levels chosen for this example are the following:

- zero--No proposals from the research category are funded.
- minimal--The least cost set of proposals that provides a modest step toward the goal. Usually, a dollar amount is set to limit the cost of the package and to constrain the choice of proposals included.
- recommended--A set of proposals that, if successfully completed, would result in significant progress toward the goal.
- enhanced--A set of proposals that, if successfully completed, would result in the major objectives for the category being met.

After ranking the individual proposals and ordering them on their value-to-cost ratios, the evaluators have the information at hand necessary to generate the recommended funding packages for the different levels of funding. Having already evaluated the individual proposals, the evaluators are very familiar with specific research benefits that would result from their successful completion. Thus, they know which proposals are duplicating research, which proposals are complementary, or which are superadditive in meeting research needs. The evaluators also have in summary form the budgetary requirements of the proposals and their value-to-cost scores. With all of this information, the evaluators are in an excellent position to recommend combinations of proposals for different levels of funding within a given category. At this stage, proposals with marginal merit or proposals presented by institutions or organizations with questionable qualifications can be dropped from further consideration. It should also be pointed out that a package that represents a higher level of funding within a particular category does not necessarily include all of the proposals recommended for the next lower-level set. This is because, at a higher funding level, it may be possible to fund a combination of more valuable research but only by excluding some proposals that may have been funded at a lower level.

Each funding package is subsequently treated as a "proposal." Therefore, some description of how the funding package addresses the evaluation criteria will be needed. As a minimum, recommended funding packages must include the list of proposals to be funded and the total cost of funding. How much additional information the packages should contain will depend on whether all evaluators rated all proposals and created all the funding packages and, consequently, would be familiar with what each package would provide. If evaluators are split along research categories, then it will be mandatory that some description be given of the benefits of funding categories at each of the levels, and it may be valuable to provide such a description in either case. Just as in the case with proposals, it was necessary to present all the information necessary for their evaluation on the criteria for which they were to be scored; so too, these package descriptions need to provide a basis for their subsequent evaluation. Thus, thought needs to be given beforehand to the criteria to be used for evaluation of combinations of proposals. The end product of this task should be a list of combinations of proposals to fund for various funding levels and a description of the benefits for each of the different levels.

3.5.4 Prioritizing Research Programs

The next step in the research prioritization process is to evaluate the recommended funding packages. Each of these packages needs to be scored on the criteria developed for this purpose. These criteria could possibly be the same as those used to evaluate individual proposals; however, it is more likely that they will be some variant of those criteria. For example, qualifications of researchers and institutions would probably not be an appropriate criterion at this level of evaluation. The reason is that there would probably be too many of such institutions in a particular package, and the questionable or poor performers will have already been screened out. Also, it may be necessary to add new criteria that deal with decisions at this higher level of evaluation. For example, the compatibility of the research program across the various categories may become a consideration at this level. Most likely, the criteria will mainly focus on the same technical concerns that were developed for evaluating individual proposals.

New weights must be established for the criteria used to evaluate the funding packages. The swing-weighting process described above is the recommended method for arriving at the weights. If the criteria contain, as a subset, some of the same criteria used to evaluate the individual proposals, then it will be possible to obtain the relative weights of those criteria from the earlier work. In any case, the fact that the evaluators have already been through the exercise and are familiar with many of the scales and their end points should facilitate the process.

Scoring, tabular reporting, and summarizing the results are identical to the proposal-rating process described above (see Tables 3.10, 3.11, and 3.12). However, Table 3.13 summarizes the results of the evaluation of the funding packages in a slightly different way. The first column lists the research categories, next is the column for the overall evaluation, then a column for the cost. The next two columns show the marginal value (ΔV) and marginal cost (ΔC). The marginal value is the increase in value that would occur for funding at that level rather than the next lowest level. The marginal cost is the marginal increase in cost for that level of funding over the next lowest level. The final column is the marginal value to marginal cost ratio ($\Delta V/\Delta C$). As with the summary sheet for the individual proposals, the information is duplicated and sorted from highest to lowest ratio. Finally, a cumulative cost is calculated based on this sequence.

3.5.5 Value-Based Planning

The final step in the research prioritization process is to determine the optimum combination of funding across the various research categories subject to the overall budgetary constraints. In this example, each of the four categories can be funded at one of four funding levels, so there are 256 possible allocations. Only 12 of these are "efficient" allocations that maximize value per dollar spent. These are determined from the marginal-value-to-marginal-cost ratios (i.e., $\Delta V/\Delta C$). Funding packages were ordered, based on these ratios, and the cumulative dollar amount was calculated. The efficient allocation is to fund projects in the order that maximizes marginal-value-to-marginal-cost ratios until the budget is expended. Thus, in this example, for a budget of $1.41M, the Clouds, Aerosols, and Radiation and

TABLE 3.13. Comparison of Research Programs

FUNDING PACKAGE:					
	VALUE	COST(10K)	ΔV	ΔC(10K)	ΔV/ΔC
CLOUDS, AEROSOLS, RADIATION					
C-A-R:ZERO	0	0	0	0	
C-A-R:MINIMUM	45	51	45	51	0.88
C-A-R: RECOMMENDED	65	75	20	24	0.83
C-A-R: ENHANCED	80	115	15	40	0.38
CHEMISTRY					
CHEM: ZERO	0	0	0	0	
CHEM: MINIMUM	35	41	35	41	0.85
CHEM: RECOMMENDED	55	65	20	24	0.83
CHEM: ENHANCED	75	95	20	30	0.67
METEOROLOGY					
MET:ZERO	0	0	0	0	
MET:MINIMUM	40	52	40	52	0.77
MET:RECOMMENDED	55	75	15	23	0.65
MET:ENHANCED	70	108	15	33	0.45
NUMERICS					
NUM:ZERO	0	0	0	0	
NUM:MINIMUM	20	35	20	35	0.57
NUM:RECOMMENDED	35	68	15	33	0.45
NUM:ENHANCED	40	82	5	14	0.36
		CUM			
		COST(10K)	ΔV	ΔC(10K)	ΔV/ΔC
C-A-R:MINIMUM		51	45	51	0.88
CHEM: MINIMUM		92	35	41	0.85
C-A-R: RECOMMENDED		116	20	24	0.83
CHEM: RECOMMENDED		141	20	25	0.80
MET:MINIMUM		193	40	52	0.77
CHEM: ENHANCED		223	20	30	0.67
MET:RECOMMENDED		246	15	23	0.65
NUM:MINIMUM		281	20	35	0.57
MET:ENHANCED		314	15	33	0.45
NUM:RECOMMENDED		347	15	33	0.45
C-A-R: ENHANCED		387	15	40	0.38
NUM:ENHANCED		401	5	14	0.36

Chemistry categories would both be funded at their recommended levels. Neither Meteorology nor Numerics would be funded.

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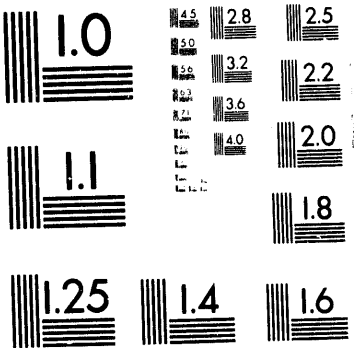
APPENDIX A

CURRENT STATUS OF GLOBAL-SCALE CHEMICAL TRANSPORT MODELS


1.0
2.8
2.5
3.2
2.2
3.6
4.0
2.0
1.1
1.8
1.25
1.4
1.6

2 of 2

APPENDIX A

CURRENT STATUS OF GLOBAL-SCALE CHEMICAL TRANSPORT MODELS

Several chemical transport models (CTMs) for the global scale currently exist. These models provide the starting point for the design of the global tropospheric chemistry modeling system. An analysis of the process modules comprising these models is presented in Table A.1. By adding other options for each module to this table, a shell for designing a modular system that incorporated not only the existing practice for each module but also included new options that embody the direction of scientific development was created.

Most of the three-dimensional CTMs for the global scale are under continual improvement and several other newer CTMs are currently under development at various locations (e.g., the National Center for Atmospheric Research and Lawrence Livermore National Laboratory). In the past, three-dimensional, global-scale models either treated only tropospheric chemical processes or stratospheric processes. Currently, many formerly stratospheric-only models are being extended down into the troposphere, and tropospheric models are being extended into the lower stratosphere. Therefore, it is difficult to provide a completely up-to-date description of the current state of the science in global-scale chemical models because this is an extremely active area of atmospheric science research. In the following, a brief synopsis of several global-scale, three-dimensional tropospheric chemistry models that have an established record of published investigations is provided.

A.1 GEOPHYSICAL FLUID DYNAMICS LABORATORY

The Geophysical Fluid Dynamics Laboratory (GFDL) CTM was developed by Mahlman et al. (1980) to study the distribution of tropospheric O_3 (Levy et al. 1985), the distribution and variability of N_2O (Levy et al. 1982; Mahlman et al. 1986), and reactive nitrogen chemistry and transport (Levy and Moxim 1987; Levy and Moxim 1989a, 1989b; Moxim 1990; Kasibhatla et al.

TABLE A.1. Features of Current Chemical Transport Models for the Global Scale

Module	Geophysical Fluid Dynamics Laboratory	Harvard University
CHEMISTRY		
Gas phase	CH_4/CO, NO_x/O_3, performed off-line	CH_4/CO, mult. lumped HC, NO_x/O_3
Aqueous phase	none	none
Aqueous size	none	none
Subgrid	instant dispersion	plume in grid
Aerosols	bulk/spec. int. mxd.	bulk/spec. int. mxd.
DYNAMICS		
Boundary layer/turbulence	K-theory, 1st order K_{zz}	K-theory, 1st order K_{zz}
Stratosphere	dynamic strat./tropo. exchange with chemistry (domain extends into lower strat.)	dynamic strat./tropo. exchange with chemistry (domain extends into lower strat.)
CLOUD PROCESSES		
Microphysics	bulk	bulk
Scavenging	scavenging coefficients	scavenging coefficients
Subgrid	fractional area	fractional area
Activation	none	none
Convective clouds	enhanced K_{zz} in moist unstable environments	decoupled/Jacob et al. (1993a)
SURFACE EXCHANGE	constant, species dependent	resistance methodology
ACTINIC FLUX		
Radiative transfer	look-up table	look-up table
Subgrid	uniform	uniform
NUMERICS		
Transport	finite volume method	moments (Prather 1986)
Chemistry	look-up table	polynomial fit to off-line model runs
Grid	uniform	uniform
SENSITIVITY/UNCERTAINTY	none	none

TABLE A.1. (contd)

Module	MOGUNTIA (NO_x/O_3 or S version)	Los Alamos National Laboratory
CHEMISTRY		
Gas phase	NO_x/O_3 and CO/CH_4 or S reactions with OH fields from NO_x/O_3 version	monthly averaged OH and $O(^1D)$ from a photochemical box model with prescribed NO_x, O_3, CO, HNO_3, and $(H_2O)_v$
Aqueous phase	none or sulfur reactions	none
Aqueous size	none or bulk	none
Subgrid	instant dispersion	instant dispersion
Aerosols	bulk/spec. int. mxd.	none
DYNAMICS		
Boundary layer/turbulence	K-theory, 1st order K_{zz}	K-theory, 1st order K_{zz} from GCM
Stratosphere	dynamic strat./tropo. exchange with chemistry (domain extends into lower strat.)	model includes strat.
CLOUD PROCESSES		
Microphysics	bulk	none
Scavenging	scavenging coefficients	none
Subgrid	fractional area	none
Activation	none	none
Convective clouds	decoupled/Feichter and Crutzen (1990)	none
SURFACE EXCHANGE	constant, species dependent	none
ACTINIC FLUX		
Radiative transfer	look-up table	look-up table
Subgrid	uniform	uniform
NUMERICS		
Transport	1-D finite difference method	1-D finite element for horizontal, 1-D finite difference for vertical
Chemistry	no published information	nonstiff
Grid	uniform	uniform
SENSITIVITY/UNCERTAINTY	none	none

TABLE A.1. (contd)

Module	GRANTOUR	Pacific Northwest Laboratory
CHEMISTRY		
Gas phase	Lurmann et al. (1986)	specified OH
Aqueous phase	none	sulfur rxns
Aqueous size	none	bulk
Subgrid	none	none
Aerosols	bulk single species	bulk/spec. int. mxd.
DYNAMICS		
Boundary layer/turbulence	K-theory, 1st order K_{zz}	K-theory, 1st order K_{zz}
Stratosphere	GCM resolved with fixed strat. O_3 or Chapman	boundary cond.
CLOUD PROCESSES		
Microphysics	none	bulk
Scavenging	scavenging coefficients	bulk
Subgrid	fractional area (stratiform, convective)	fractional area
Activation	bulk parameterization	bulk activation
Convective clouds	coupled conv. adjustment	decoupled/Berkowitz et al. (1989)
SURFACE EXCHANGE	prescribed deposition velocities	constant, species dependent
ACTINIC FLUX		
Radiative transfer	look-up table	look-up table
Subgrid	uniform	uniform
NUMERICS		
Transport	Lagrangian	1-D finite-difference method
Chemistry	Sillman (1991)	new solver
Grid	uniform	uniform
SENSITIVITY/UNCERTAINTY	none	none

1991, 1993). The most recently published version of the GFDL CTM (Kasibhatla et al. 1993) used a horizontal resolution of 265 km and 11 vertical levels from the surface to 10 mb. Wind and precipitation fields, time-averaged over 6-h intervals, were obtained from the GFDL general circulation model (Manabe et al. 1974; Manabe and Holloway 1975) over a 12-month period. The GFDL CTM transports three species (NO_x, HNO_3, and peroxyacetyl nitrate) and uses production and loss rates for these species calculated off-line but carried in the model as temporally varying, two-dimensional fields. A parameterized wet removal process is used to simulate the precipitation scavenging of HNO_3, whereby the amount of HNO_3 removed from a particular grid box is a function of the local precipitation rate and is proportional to the local HNO_3 mixing ratio. The parameterization differentiates between shallow and deep convective events. Model inputs to the off-line chemical module include hemispherically averaged one-dimensional profiles of CO and NO_x and two-dimensional profiles of CH_4, O_3, water vapor, temperature, and total column O_3.

A.2 HARVARD UNIVERSITY

Harvard University's global-scale CTM has been used to simulate the distributions of several trace atmospheric species, including chlorofluorocarbons (Prather et al. 1987), ^{85}Kr (Jacob et al. 1987), CH_3CCl_3 (Spivakovsky et al. 1990a, 1990b), ^{222}Rn (Jacob and Prather 1990; Balkanski and Jacob 1990; Balkanski et al. 1992), and ^{210}Pb (Balkanski et al. 1993). In sub-hemispheric versions, the model has been used to simulate the production and export of O_3 from North America (Jacob et al. 1993a, 1993b). The most recently published version of the model (Jacob et al. 1993a) solves mass conservation equations for six species (odd oxygen, NO_x, peroxyacetyl nitrate, CO, a short-lived hydrocarbon, and a long-lived hydrocarbon) on a domain with 4-degree latitude x 5-degree longitude horizontal resolution and 9 vertical layers from the surface to 10 hPa. Meteorological data used to drive the CTM are derived from archives of the Goddard Institute for Space Studies general circulation model (Hansen et al. 1983) with 4-h averages of wind fields, mixing depths, and convective mass fluxes and 5-day averages of temperature, humidity, and cloud reflectivities. The chemical mechanism of Lurmann et al.

(1986) is parameterized into a set of polynomials that describe the chemical environment in terms of the six computed species (Spivakovsky et al. 1990a). The transport numerics of the model are as described by Prather (1986) and tested by Jacob et al. (1989). Subgrid-scale plumes are treated via the nested plume method of Sillman et al. (1990).

A.3 MOGUNTIA

The MOGUNTIA model (Zimmerman 1988; Zimmerman et al. 1989) was developed at the Max Planck Institute for Chemistry in Mainz, Germany, and has been used to study global $CFCl_3$ and CH_3CCl_3 distributions (Zimmerman 1988), ^{85}Kr distributions (Zimmerman et al. 1989), ^{222}Rn distributions and vertical transport (Feichter and Crutzen 1990), changes in global oxidative capacity (Crutzen and Zimmerman 1991), and heterogeneous atmospheric processes (Dentener and Crutzen 1993). Another implementation of the MOGUNTIA model has been designed for the study of the global sulfur cycle (Langner and Rodhe 1991). Horizontal resolution of the model is 10 degrees latitude x 10 degrees longitude, while the vertical domain extends from the surface to 100 hPa with 10 vertical layers. Large-scale transport is described by monthly averaged winds (Oort 1983), while convective cloud transport is parameterized according to Feichter and Crutzen (1990). The chemical transformations included in MOGUNTIA account for $CH_4/CO/NO_x/HO_x$ photochemistry in remote areas, as described in Crutzen and Zimmerman (1991). The sulfur version of the model uses monthly averaged OH fields from the NO_x/O_3 version to describe the gas-phase oxidation of SO_2 and dimethylsulfide. Aqueous-phase sulfur oxidation is treated in a highly parameterized manner, based on average cloud lifetimes (Langner and Rodhe 1991).

A.4 LOS ALAMOS NATIONAL LABORATORY

Los Alamos National Laboratory's chemical tracer model (Tie et al. 1991) has been used for multiyear simulations of long-lived species, including CH_4 (Tie et al. 1991), CFC-11 (Kao et al. 1992), and CH_3CCl_3 (Tie et al. 1992). The model accounts for emissions, advective and diffusive transport, and limited gas-phase chemistry. Meteorological data to drive the CTM simulations

are derived from the Los Alamos general circulation model (Kao et al. 1990). Oxidant concentrations for the three-dimensional model are precalculated using an off-line two-dimensional (latitude-altitude) model that employs prescribed latitude-altitude distributions of NO_x, O_3, CO, HNO_3, and water vapor. The horizontal resolution of the model is 4.5 degrees latitude x 7.5 degrees longitude, while the vertical domain extends from the surface to approximately 9 hPa and employs 20 levels.

A.5 GRANTOUR

The GRANTOUR model was developed at the Lawrence Livermore National Laboratory (Penner et al. 1993). The distinctive feature of GRANTOUR is its use of Lagrangian air parcels to account for advection of constituents, which minimizes spurious dispersion and permits advection of a large number of species at once. The air parcels are assumed to be constant mass, which implies poor representation of vertical structure in the stratosphere. At least three versions of GRANTOUR have been developed. One focuses on O_3 and nitrogen chemistry, using the Lurmann et al. (1986) mechanism (Penner et al. 1991a). A second version simulates the sulfur cycle, using prescribed OH concentrations and parameterized scavenging (Erickson et al. 1991). Aqueous-phase chemistry is neglected. Sulfate mass concentration is related to aerosol number density by assuming a constant dry size, and direct scattering of sunlight is calculated using the Kohler solution of size dependence on relative humidity. A third version treats the emission, transport, and removal of biomass smoke and soot particles (Penner et al. 1991b, 1992). GRANTOUR is usually driven by winds, temperature, humidity, convective mass flux, and precipitation simulated by the Lawrence Livermore version of the National Center for Atmospheric Research's Community Climate Model.

A.6 GLOBAL CHEMISTRY MODEL

The Global Chemistry Model (GChM) was developed at Pacific Northwest Laboratory to study the global-scale reaction, redistribution, and wet removal of soluble sulfur species (Luecken et al. 1991). Recently, another version of the GChM has been developed to study the global distribution of CO (Easter

et al. 1993). The model is designed to utilize meteorological information from either observational sources or general circulation models and to account for the vertical redistribution of pollutants resulting from convective cloud transport via a parameterization as described in Luecken et al. (1991). The sulfur version of the GChM (Luecken et al. 1991) accounts for the gas- and aqueous-phase oxidation of SO_2 and dimethylsulfide through parameterized oxidation rates. Both stratiform and convective clouds are included in the model. Each provides vertical profiles of cloud water, which in turn, allows for an explicit treatment of aqueous-phase sulfur chemistry. The CO/CH_4 version of the GChM (Easter et al. 1993) calculates the transformation rates of CO and CH_4, based on the gas-phase mechanism of Lelieveld and Crutzen (1991). By prescribing global distributions of NO_x and O_3, and assuming a pseudo-steady state for all species in the mechanism except CO and CH_4, the computational burden of the chemical calculations is reduced. This version of the GChM has been used to analyze midtropospheric CO data from the National Aeronautics and Space Administration's Measurement of Air Pollution by Satellites Program (Easter et al. 1993).

A.7 TWO-DIMENSIONAL GLOBAL MODELS

Several two-dimensional global atmospheric chemistry models have been developed over the past 15 years (Isaksen and Rodhe 1978; Derwent 1982; Crutzen and Gidel 1983; Hough 1989, 1991; Law and Pyle 1993a, 1993b). Although two-dimensional models are very useful tools for general analysis of many atmospheric processes and species (Isaksen and Rodhe 1978; Rodhe and Isaksen 1980; Derwent 1982; Crutzen and Gidel 1983; Isaksen and Hov 1987; Hough 1989, 1991; Kanakidou et al. 1991; Law and Pyle 1993a, 1993b), their usefulness is limited for the complex assessment and policy formulation activities that will be necessary over the next several decades. The social and economic changes that are likely to happen in the next century will be spatially nonuniform. As industrial development in South America, Africa, and Asia continues to accelerate, the atmospheric impacts of their growth will not be adequately modeled by zonally averaged two-dimensional models. As radiatively important trace species emissions from these areas will be

distributed nonuniformly in both space and time and as shorter-lifetime species become more important in the global radiative balance, three-dimensional atmospheric chemistry models become the most appropriate tool for detailed policy assessment and analysis.

A.8 OTHER EFFORTS

As mentioned before, global tropospheric chemistry modeling is currently a very active area of research. Many groups are presently developing capabilities for three-dimensional simulation of global tropospheric chemical processes. The above brief discussions focus on entities that have published studies involving their models in refereed journals. At a recent workshop on global tropospheric chemistry modeling (NASA 1993), several groups having some level of development currently in progress in this area were represented. These groups included the National Center for Atmospheric Research, Cambridge University, Massachusetts Institute of Technology, Atmospheric and Environmental Research, Inc., Goddard Institute for Space Studies, and Lawrence Livermore National Laboratory. Some of these efforts are being built on two-dimensional atmospheric chemistry models (National Center for Atmospheric Research, Lawrence Livermore National Laboratory, Atmospheric and Environmental Research, Inc.), while others are being derived from tracer implementations of general circulation models (National Center for Atmospheric Research, Goddard Institute for Space Studies). Still others are being derived from stratospheric chemistry models, extending their domain down into the troposphere (National Center for Atmospheric Research). Because all of these efforts are in relatively early developmental stages, little or no published information is available on their structure or applicability. It is indeed conceivable that some of these efforts will be abandoned in the near future as competition in this research area increases. It is also conceivable that one of these efforts or one of the more established models may form a suitable basis for the global tropospheric chemistry modeling system as described in this report.

A.9 REFERENCES

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APPENDIX B

DESIGN OPTIONS FOR A GLOBAL TROPOSPHERIC CHEMISTRY MODEL

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DESIGN OPTIONS FOR A GLOBAL TROPOSPHERIC CHEMISTRY MODEL

B.1 MODULE OPTIONS: A DESIGN MENU

The design menu (Table B.1) of module options was formulated to facilitate the construction of a design vision for a global tropospheric chemistry model. As far as possible, each module that would exist within the model is represented by an underlined heading. Under each of these modules an attempt was made to list all possible options that a model designer might have in constructing that module. Each of these modules was then grouped into the broader categories of 1) chemistry, 2) dynamics, 3) cloud processes, 4) surface exchange, 5) actinic flux, 6) numerics, and 7) sensitivity/uncertainty. To design a version of the model, one chooses appropriate options from each module, keeping in mind that some combinations of options would not be good overall designs. For example, choosing a size-resolved/species-resolved treatment of aerosols would not be appropriate with only a bulk treatment of cloud microphysics and aerosol activation. Thus, the design menu serves as an organizing tool for an experienced model designer and would not be useful for persons with limited atmospheric chemistry modeling experience. The following is an expanded description of the possible choices for each module in the design menu.

B.1.1 Chemistry

B.1.1.1 Gas Phase

In this module, a wide variety of possible combinations of included chemical reactions and approaches exists. Each of the individual options is described below:

- NO_x/O_3--basic NO_x/O_3 photochemistry, including nighttime NO_x conversion
- CH_4/CO--chemistry for the tropospheric oxidation of CH_4 to CO

TABLE B.1. Module Options for a Global Tropospheric Chemistry Model

Chemistry

Gas phase	Aqu. phase	Aqu. Size	Subgrid	Aerosols
_CH_4/CO	_none	_none	_instant dispersion	_none
_3 lumped HC	_sulfur rxns	_bulk	_plume in grid	_bulk/spec. int. mxd.
_mult. lmpd HC	_radical rxns	_moments	_statistical correlation	_bulk/spec. resolved
_explicit HC	_Cl rxns	_size resolved		_moments/spec. int. mxd.
_hybrid	_$NO_y/O_3/H_2O_2$			_moments/spec. resolved
_halogens				_size-resolved/species internally mixed
_NO_x/O_3				_size-resolved/species resolved
_sulfur rxns				
_NH_3				
_specified OH				

Dynamics

Boundary Layer/Turbulence	Stratosphere
_K-theory, 1st order K_{zz}	_boundary cond.
_K-theory, 1.5 order K_{zz} (TKE)	_dynamic strat/tropo exchange w/o chem
_K-theory, 2nd order K_{zz}	_dynamic strat/tropo exchange w/ chem
_Second order for chemistry and meteorology	
_Nonlocal closure	

Cloud Processes

Microphysics	Scavenging	Subgrid	Activation	Convective Clouds
_none	_none	_uniform	_none	_none
_bulk	_scav coeff	_fract area	_bulk activation	_decoupled/Walcek&Taylor
_moments	_bulk	_statistical	_moments activation	_decoupled/Berkowitz et al.
_size resolved	_moments	_cld dist from GCM	_size-resolved activation	_coupled/convective adj.
_ice	_size resolved			_coupled/Kuo scheme
				_coupled/Arakawa-Schubert
				_coupled/"3rd generation"

Surface Exchange

_constant, species dependent
_resistance methodology
_surface exchange model

Actinic Flux

Radiative Transfer	Subgrid
_look-up table	_uniform
_on-line cloud effects	_cloud fraction
_on-line radiative transfer calcs.	_statistical 1-D
	_3-D effects

Numerics

Transport	Chemistry	Grid
_lagrangian (GRANTOUR)	_Sillman	_uniform
_1-D FDM	_QSSA	_irregular
_1-D FEM	_hybrid	_adaptive
_2-D FEM	_Gear	
_moments (Prather)	_new sol	
	_non-stiff solver	

Sensitivity/Uncertainty

_none
_Monte Carlo
_direct decoupled
_adjoint
_stochastic variables
_automatic differentiation

- 3 lumped HC--chemical reactions for two lumped surrogate anthropogenic species and one biogenic species
- mult. lmpd HC--chemical reactions for lumped hydrocarbon (HC) species for the major classes (e.g., alkanes, alkenes, aldehydes, aromatics, etc.)
- explicit HC--chemical reactions for all important HC species included and represented explicitly
- sulfur rxns--chemical reactions for the oxidation of SO_2 and dimethylsulfide
- halogens--chemical reactions of Cl and Br species in the troposphere
- NH_3--chemical reactions of NH_3
- hybrid--a technique whereby simple background chemistry is used in remote areas, while a more detailed mechanism that includes HC is employed in polluted areas
- specified OH--techniques whereby hydroxyl radical concentrations are specified through an empirical parameterization or by the specification of precursor species.

B.1.1.2 Aqueous Phase

As in the gas-phase module, several possible combinations of aqueous-phase chemical treatments are possible. The individual options are described below:

- none--no aqueous-phase reactions are included
- $NO_y/O_3/H_2O_2$--basic $NO_y/O_3/H_2O_2$ dissolution and equilibria
- radical rxns--aqueous-phase radical dissolution, equilibria, and reactions (e.g., aqueous-phase mechanism of Lelieveld and Crutzen 1991)
- sulfur rxns--aqueous-phase oxidation reactions of S(IV) species to S(VI) species
- Cl rxns--aqueous-phase Cl dissolution, equilibria, and reactions.

B.1.1.3 Aqueous-Phase Size Dependence

Recent research has indicated that different-size aqueous droplets may have significantly different species concentrations and rates of reaction.

This module accounts for the possibility of various treatments of aqueous-phase size dependence. Either a bulk, moments, or size-resolved treatment may be chosen in this category. The bulk treatment assumes one size droplet for each aqueous phase, while the size-resolved treatment employs several discrete size bins for each aqueous phase. A moments treatment would require the specification of a distribution of droplet sizes for each aqueous phase with associated chemical differences.

B.1.1.4 Subgrid

This category refers to the treatment of species emissions as they are introduced into the model grid. Either instantaneous dispersion, plume-in-grid, or statistical correlation treatments may be chosen in this category. In most current regional-scale models, emissions within a grid cell are lumped, emitted, and instantaneously mixed throughout the local grid cell volume. Of course, in the real situation, plumes often can retain a coherent identity over scales that span a typical horizontal grid cell spacing. Significant differences can then occur in the representation of chemical transformations that occur within those plumes as compared to the reduced reaction rates that occur as a result of instantaneous dispersion. Some modelers have, therefore, devised schemes that explicitly calculate plume concentrations from major sources located in the subgrid domain and translate them to concentrations that occur at the model's grid nodes. Another possible technique to account for subgrid chemical behavior is the statistical correlation method, whereby each species is represented by a subgrid statistical distribution. Using the species distributions, cross correlations can be calculated and incorporated into subgrid chemical calculations.

B.1.1.5 Aerosols

Treatment of aerosols involves both their physical size distribution and their chemical composition. There are three options for the physical size. The bulk treatment only predicts the aerosol volume, and no size information is provided. (Actually, the mass of different chemical components in the aerosol would be predicted, such as sulfate, elemental carbon, organics, etc.) The size-resolved treatment predicts the size distribution by dividing the aerosol population into a set of size bins, but is computationally expensive.

The moments method assumes that each mode of the aerosol population (nuclei, accumulation, coarse) can be represented by a log-normal size distribution. The log-normal distribution is determined by three parameters that the model predicts. The moments method is computationally less expensive than the size-resolved method but also less rigorous.

B.1.2 Dynamics

B.1.2.1 Boundary Layer and Turbulent Exchange

The K-theory, or first-order closure approach for turbulent mixing, which is used in most atmospheric chemistry models, parameterizes the vertical turbulent flux of a species in terms of the vertical gradient of the species. This approach requires obtaining a vertical exchange coefficient from the meteorological driver model. The meteorological driver itself will have some turbulence parameterization (first-order closure; one- and one-half-order closure, which is called the turbulent kinetic energy approach; or second-order closure). Note that the parameterization used in the meteorological driver influences the values but not the form of the vertical exchange coefficients. Another approach is to use second-order closure in both the chemistry and meteorological models. This requires simulation of both mean concentrations and turbulent fluctuations of concentrations and, to date, has not been used in any operational model. In both the first- and second-order closure approaches, material is transported from one layer to an adjacent layer and then mixes throughout that layer. The nonlocal closure approach recognizes that eddies responsible for turbulent transport may move across several vertical layers before they stop and dissipate. Mathematically, turbulent transfer can occur between every pair of layers in the model. The basis for determining the exchange rates is less well developed than K-theory.

B.1.2.2 Stratospheric Exchange

Mass exchange between the troposphere and stratosphere is a very important process for many species in tropospheric chemistry. A significant source of O_3 and NO_y in the troposphere is thought to be transported from the stratosphere. The upper boundary of the model could be placed either at the tropopause or within the lower stratosphere. A simple boundary condition can be

applied in either situation, or with the upper boundary within the stratosphere, dynamic exchange can take place across the tropopause, based on transport quantities provided by the driving general circulation model. With the upper boundary within the lower stratosphere, chemistry could be calculated explicitly or a chemical climatology could be specified.

B.1.3 Cloud Processes

The treatment of cloud processes, such as microphysics, aerosol activation and scavenging, and vertical transport, offers a variety of options. Microphysics can be neglected entirely for insoluble species. For most chemical species, cloud microphysics must be represented in some manner. For a comprehensive treatment of clouds, the size distributions of cloud variables and cloud processes would be explicitly resolved into a number of size bins. For many purposes, a bulk approach is acceptable, in which only the mass of cloud water and precipitation is allowed to vary. A more general approach considers variations in other moments of the cloud particle-size distribution; variations in droplet number concentration are of particular relevance to aerosol interactions. One may wish to distinguish the ice from the liquid phase for certain applications.

The treatments of aerosol activation and scavenging largely follow the representation of the cloud and aerosol variables. If the bulk approach is used for clouds and aerosols, it should be used for activation and scavenging. If the size distribution of cloud particles and aerosols is explicitly resolved, so could be the treatment of activation and scavenging. If cloud microphysics is not treated, scavenging could still be treated using the simulated or observed precipitation rates and prescribed scavenging coefficients.

Subgrid-scale variations in cloud processes can either be neglected entirely, expressed in terms of a parameterized, homogeneous, cloudy grid cell fraction, or treated using idealized probability distributions for the cloud variables, in which the parameters of the probability distributions are related to a few predicted moments of the probability distributions. A final option applicable for a model that is driven by the meteorological fields from a general circulation model is to use the clouds' fields derived within that model.

Vertical transport of soluble and insoluble species by convective clouds can be treated using the off-line schemes of Walcek and Taylor (1986) or Berkowitz et al. (1989). Other optional schemes are derived from general circulation models; namely, the moist convective adjustment, the Kuo scheme, or the Arakawa and Schubert (1974) parameterization. Each of these schemes has shortcomings, so that one might consider developing a "third generation" convective parameterization. It should be recognized that, because each scheme is based on different equilibrium assumptions, the scheme selected for the model should be the same as that used in the host general circulation model; otherwise, highly inconsistent vertical transport is diagnosed.

B.1.4 Surface Exchange

Chemical exchange between the atmosphere and land or ocean surface will be an important aspect of the model. This exchange can be implemented through temporally constant, species-dependent surface fluxes, based on empirical information or through the resistance analogy methodology summarized by Wesely (1989). A more sophisticated surface-exchange model could also be developed, wherein dynamic fluxes of material may be exchanged in either direction, based on the currently prevailing chemical and meteorological conditions. The construction of the surface-exchange model could benefit greatly from research carried out in the agricultural and oceanographic communities.

B.1.5 Actinic Flux

Actinic flux depends on the solar zenith angle, the profile of trace chemical concentrations, the surface reflectivity, and the distribution of clouds. Actinic flux has in the past been evaluated using look-up tables for a predetermined combination of solar zenith angle, surface reflectivity, and chemical and cloud profiles. Given the large variability of clouds and the weak wavelength dependence of scattering by clouds, one option would be to retain the look-up approach for the clear-sky actinic flux for each absorption line, but apply an on-line modification factor associated with the influence of clouds on the actinic flux profile. A third option is to abandon look-up tables entirely; instead, calculating the full radiative transfer for each absorption band. Subgrid cloud effects could be neglected or treated using the simple homogeneous cloud fraction approach (with various cloud overlap

assumptions) or a more general statistical description of the subgrid distribution of cloud optical properties.

B.1.6 Numerics

B.1.6.1 Transport

Several numerical techniques are currently used in regional- and global-scale atmospheric chemistry models for the solution of the species transport equations. The possible choices listed in this category include 1) the Lagrangian parcel technique of Walton et al. (1988); 2) the second-order moments method of Prather (1986); 3) time splitting of the three-dimensional equation into several one-dimensional equations, which are then solved by finite-difference methods (e.g., Bott 1989); 4) time splitting with the one-dimensional equations solved by finite-element methods (e.g., Galerkin methods); or 5) time splitting of the three-dimensional equation into a two-dimensional horizontal transport equation and a one-dimensional vertical equation, which are then solved by finite-element methods (this technique allows for the possibility of irregular or adaptive grid techniques).

B.1.6.2 Chemistry

The solution of the system of ordinary differential equations that describe the chemical transformations of modeled species is the most computationally intensive portion of atmospheric chemistry models. Consequently, the choice of the ordinary differential equation solver for these calculations is critically important. Several techniques are currently used, including the methodology of Sillman (1991), which is limited to tropospheric chemical simulation only; the quasi-steady-state approximation methodology of Hesstvedt et al. (1978), which is used extensively by current regional-scale models; the hybrid predictor-corrector methodology of Odman et al. (1992), which is also used by several current regional-scale models; or the traditional Gear methodology. Also included as choices in this category are a new solver, which does not exist currently, but is wished for by all atmospheric chemistry modelers, and a simple nonstiff solver, which would be used only if CH_4/CO chemistry and specified OH were chosen under the gas-phase module.

B.1.6.3 Grid

A uniform horizontal grid (possibly with a nonuniform vertical grid) has been the predominant type of grid structure used in the past for regional- and global-scale models. Only recently (Mathur et al. 1992) have irregular grid structures been employed in regional-scale models. The purpose of irregular grids is to specify a priori a grid structure that has greater resolution in areas where steep concentration gradients are expected or where subgrid chemical effects may be the largest (i.e., near major sources). For a global model, an irregular grid application would most likely result in a finer grid over continental areas and a much coarser grid over ocean areas. Adaptive grid techniques have been used for many years in advanced computational fluid dynamics calculations (e.g., shock wave propagation), but have not been used in atmospheric chemistry models to date. In the adaptive grid methodology, an irregular grid is used and modified dynamically during run time, based on changing concentration gradients or meteorological features. Adaptive grid techniques, though extremely powerful in the efficient computation of a highly accurate solution, are computationally expensive. Of course, the grid structure that is ultimately chosen must be closely tied to the numerical technique chosen for solution of the transport equations. Although a uniform grid can be used with most numerical techniques, this is not true for irregular or adaptive grids. The most straightforward application of the irregular or adaptive grid techniques comes with the use of multidimensional finite-element techniques.

B.1.7 Uncertainty/Sensitivity

A full discussion of the various approaches to uncertainty and sensitivity analysis is given in the text of this report. There will be no one module within the model that handles uncertainty or sensitivity. Rather, the means for obtaining information on uncertainty or sensitivity must be built into individual modules or be determined by manipulating the model after it is completed. It is recommended that approaches to uncertainty and sensitivity estimation be built into the modules and model as the codes are being developed.

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APPENDIX C

RESEARCH ACTIVITIES DEFINED AT THE SECOND WORKSHOP

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Small groups were convened to discuss the research that would be required to develop the modules needed by the three classes of chemical transport models (CTMs) in the global tropospheric chemistry modeling system (GTCMS). These groups were selected according to the area of expertise of the participants. As areas in which research was needed were identified, each group produced a very brief, often sketchy description of the research activity. These unedited, hand-written descriptions were then distributed to all participants for the purpose of scoring the activities according to criteria developed at the workshop. The research activities presented in this appendix are heavily edited versions of those raw descriptions. For many activities, the specification of more than one level of effort is an added refinement not available at the time of the workshop. For the purpose of defining research, further development of the descriptions of what each research activity entails will be needed.

Research Area:	Aerosols/Nucleation
Activity Title:	Investigate Nucleation on Mixed Aerosols
Description:	Laboratory and theoretical research is needed to improve understanding of the influence of aerosol surface properties on the nucleation process. Laboratory measurements are needed to characterize aerosol surface properties sufficiently that theoretical models can be evaluated against measured nucleation supersaturation.
Estimated Level of Effort and Time:	1 person-year for 3 years (split between an experimentalist and a theoretician)

Research Area:	Clouds/Subgrid
Activity Title:	Develop Treatment of Subgrid Variability in Clouds and Cloud Processes
Description:	Subgrid-scale variability in cloud properties and cloud processes has a strong influence on soluble gases and aerosols through deposition, heterogeneous oxidation, activation, scavenging, and resuspension. A physically based treatment of subgrid cloud variability is needed to account for the influence of the variability of soluble gases and aerosols. A statistical description of subgrid variability is recommended, in which variability in cloud microphysical properties is expressed in terms of idealized probability distributions of each cloud variable. The parameters of the probability distribution are related to various statistical moments of the distribution (e.g., mean and variance), which are predicted from conservation equations. The influence of subgrid variability on cloud processes would be treated by integrating each cloud process over the probability distribution of the appropriate cloud variables. The subgrid variance should be related to subgrid variance in vertical velocity, which would be parameterized in terms of cumulus convection, three-dimensional turbulence, and mesoscale circulation.
	Level I. Develop numerical solutions to the conservation equations for the moments of the subgrid variability of cloud variables.
	Level II. Develop and test parameterizations of the subgrid variability of cloud variables that are suitable for use in a GTCMS.
Estimated Level of Effort and Time:	Level I. 1 person-year for 3 years
	Level II. 2 person-years for 3 years

Research Area: Aerosols/General

Activity Title: Develop Parameterized Moments of Species-Resolved Aerosol-Size Distributions

Description: Aerosol-size distributions can often be approximated in terms of a sum of a few log-normal distributions. This approach has already been used for single-component aerosols. However, different aerosol types arise from the diversity of aerosol sources. Although some aerosols are rather pure in composition, most are mixtures of different components. The efficient representation of mixtures of aerosol components within the context of the method of moments is a challenging research topic. Although a complete treatment of aerosol mixtures using joint-size distributions is possible, it is extremely computationally intensive. Research is needed to develop approximate representations of aerosol mixtures and their influence on aerosol processes. The nucleation, accumulation, and coarse aerosol-size modes must be distinguished. Aerosol species to be treated include H_2SO_4, $(NH_4)_2SO_4$, NH_4HSO_4, elemental carbon, crustal, and organics. Treatments of the influence of aerosol nucleation, condensation, coagulation, deliquescence, activation, resuspension, scavenging, and gravitational settling on each moment and each component are required. Note that this research activity is linked with the "Develop Moments Method for Aerosol Processing by Clouds" activity.

Estimated Level of Effort and Time: 2 person-years for 3 years

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Research Area: Aerosols/General

Activity Title: Evaluate Species-Resolved Moments Method for Aerosols

Description: Although the moments method for single-component aerosol dynamics is well established and evaluated to some extent, much remains to be done for the case of multicomponent aerosols. Multicomponent moments models need to be compared with explicit model calculations. Explicit models, in turn, need to be compared with laboratory experiments, particularly for aerosol mixtures under a variety of conditions, particularly those that isolate condensational growth and coagulation.

Estimated Level of Effort and Time: 1 person-year for 3 years

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Research Area: Aerosols/General

Activity Title: Develop Emissions Inventories of Aerosols and Their Precursors

Description: To be useful as a policy analysis tool, the GTCMS requires reliable global inventories of the different anthropogenic and biogenic sources of aerosols and aerosol precursors. Emissions inventories are needed for sulfate precursors (SO_2 and dimethylsulfide [DMS]), nitrate precursors (NO_x), NH_3, elemental carbon, organics (primary aerosol emissions and precursor gases), and mineral dust. For those emissions that depend on meteorological conditions (e.g., wind speed in the case of dust and DMS), emissions should be parameterized in terms of surface concentrations and meteorology. For other components, any seasonality in the emissions should be accounted for.

Level I. Compile existing oceanic and developing country inventories and recommend how they should be used in the GTCMS.

Level II. Improve by refining, expanding, and automating the emission inventories for the world's oceans and developing nations and provide the interface to the GTCMS.

Estimated Level of Effort and Time:

Level I. 1 person-year for 1 year

Level II. 3 person-years for 3 years

Research Area: Aerosols/Scavenging

Activity Title: Develop Moments Method for Aerosol Processing by Clouds

Description: For the aerosol model, the method of moments is recommended for the representation of both aerosols and cloud particles. Research is needed to express cloud processing of aerosols in terms of moments of both aerosols and clouds. The influence of aerosol activation, droplet coalescence, scavenging of interstitial aerosol, aqueous reactions, and resuspension on the moments of the aerosol size needs to be treated in a manner consistent with the moments representation of aerosol and cloud particle-size distributions. A complete treatment of the problem would require a joint droplet/aerosol-size distribution; a simplified treatment is required that distinguishes between activated and interstitial aerosols but does not employ the full joint-size distribution.

Estimated Level of Effort and Time: 1 person-year for 2 years

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Research Area: Radiation

Activity Title: Update Actinic Flux J-Value Look-Up Tables

Description: Photolysis rate look-up tables used in many current models were developed 10 years ago. These look-up tables should be updated to include recent information on quantum yield and absorption cross sections. Altitude effects and modification of radiation fields by clouds (on-line) should be included.

Estimated Level of Effort and Time: 0.5 person-year for 2 years

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Research Area: Chemistry/Gas Phase

Activity Title: Develop a Gas-Phase NO_x/O_3/HC Chemical Mechanism for the Short-Lived CTM

Description: Develop a gas-phase chemical mechanism that includes multiple hydrocarbon (HC) categories and appropriate secondary organic products for the short-lived species CTM. This includes appropriate selections of anthropogenic and biogenic HC species. The mechanism must be flexible to allow advances in understanding to be readily incorporated. Also, the mechanism must emphasize radical-radical interactions for low-NO_x regimes that will occur in remote areas over the globe.

Level I. Review gas-phase chemical mechanisms currently in use in global tropospheric models and make an appropriate choice for the CTM. Conduct a workshop to bring together atmospheric chemists and modelers to compile the latest understanding of gas-phase photochemistry as it would be implemented in the CTM.

Level II. Develop a gas-phase chemical mechanism that contains limited HC speciation for initial implementation and testing in the CTM.

Level III. Develop a gas-phase mechanism for the CTM that contains appropriate HC speciation and sulfur chemistry based on current laboratory and field data.

Estimated Level of Effort and Time:	Level I.	1 person-year for 1 year
	Level II.	1 person-year for 2 years
	Level III.	3 person-years for 3 years

Research Area:	Chemistry/Gas Phase	
Activity Title:	Evaluate the Effect of Grid Resolution on Subgrid NO_x Cycling and O_3 Production	
Description:	Evaluate the effect of grid resolution (and grid irregularity) on subgrid NO_x cycling and O_3 production. Determine what global grid resolution will be appropriate for the accurate prediction of subgrid behavior. Also investigate the tradeoffs between interpolating meteorological data to an irregular grid versus the loss of subgrid chemical nonlinearities on a uniform grid.	
	Level I.	Review literature on subgrid chemical nonlinearities and make appropriate choices for GTCMS implementation.
	Level II.	Perform modeling studies to evaluate the effect of uniform grid-resolution differences on subgrid NO_x/O_3 chemistry and determine appropriate parameterizations or recommendations for GTCMS implementation.
	Level III.	Perform modeling studies to evaluate the effect of uniform and nonuniform grid-resolution differences on subgrid NO_x/O_3 chemistry and determine appropriate parameterizations or recommendations for GTCMS implementation.
Estimated Level of Effort and Time:	Level I.	1 person-year for 1 year
	Level II.	1 person-year for 2 years
	Level III.	2 person-years for 2 years

Research Area:	Chemistry/Gas Phase
Activity Title:	Develop a Gas- and Aqueous-Phase Chemical Module for the Aerosol CTM

Description: Develop a gas- and aqueous-phase chemical module for the aerosol CTM, including both natural and anthropogenic sources of aerosols and precursors, such as windblown dust, biomass burning emissions, SO_2, DMS, HNO_3, NH_3, etc. This module will be significantly different from the gas- and aqueous-phase modules of the short-lived species model, in that many species important for the aerosol model will not be as important for the short-lived species model.

Level I. Review gas- and aqueous-phase chemical mechanisms in current aerosol models and make appropriate recommendations for implementation in the aerosol CTM.

Level II. Evaluate existing gas- and aqueous-phase chemical mechanisms in aerosol models, based on appropriateness for the GTCMS and the most recent laboratory and field data, and implement for the aerosol CTM.

Level III. Develop a gas- and aqueous-phase chemical mechanism for the aerosol CTM, based on the most recent laboratory and field data.

Estimated Level of Effort and Time:

Level I. 0.5 person-year for 1 year

Level II. 1 person-year for 1 year

Level III. 2 person-years for 2 years

Research Area: Chemistry/Emissions

Activity Title: Develop Emissions Inventories Required for the GTCMS

Description: Analyze the scientific needs of the short-lived, long-lived, and aerosol CTMs in terms of needed emissions inventories. Both the species required and the characteristics (e.g., temporal and spatial resolution) of the inventories as they will be applied to the different models should be included in the analysis. Emissions of sulfur compounds, NO_x, NH_3, CH_4, and other HCs by the world's oceans and from the developing nations will play a major role in global-scale chemical modeling.

Level I. Develop an interface between available emissions inventories and the GTCMS.

	Level II.	Develop or improve inventories for all the radiatively important trace species and their precursors for the world's oceans and developing nations and make compatible with the input needs of the GTCMS.
Estimated Level of Effort and Time:	Level I.	0.5 person-year for 1 year
	Level II.	2 person-years for 3 years

Research Area:	Chemistry/Aqueous Phase	
Activity Title:	Evaluate the Effect of Aerosols on Tropospheric O_3	
Description:	Investigate the impact of aerosols on the tropospheric photochemical O_3 cycle. Recent research indicates that there may be a link between photochemical O_3 production/destruction and aerosols, particularly in the marine troposphere. Examine these processes in the context of their significance for inclusion in the GTCMS. The results of this study will feed directly into the development of the short-lived species chemical mechanism and the aerosol model mechanism.	
	Level I.	Evaluate available literature on the effect of aerosols on tropospheric O_3 and make recommendations for implementation into the GTCMS.
	Level II.	Perform a model analysis of the effect of aerosols on O_3 and make recommendations for implementation into the GTCMS.
Estimated Level of Effort and Time:	Level I.	0.5 person-year for 1 year
	Level II.	2 person-years for 1 year

Research Area:	Chemistry/Aqueous Phase	
Activity Title:	Evaluate the Effect of Clouds on Tropospheric O_3	
Description:	Extend the results of recent research on the effect of clouds on tropospheric O_3 production/destruction by performing analyses that include more realistic cloud microphysics/dynamics and applying the analysis to various chemically coherent regions of the troposphere. Recent research has indicated that O_3 production/destruction may be significantly affected by aqueous-phase processes in the troposphere. The results of this study will feed directly into the development of the short-lived species chemical mechanisms.	

	Level I.	Evaluate available literature on the effect of clouds on O_3 and make recommendations for implementation in the GTCMS.
	Level II.	Perform a model analysis to evaluate the effect of clouds on O_3 and make recommendations for implementation in the GTCMS.
Estimated Level of Effort and Time:	Level I.	0.5 person-year for 1 year
	Level II.	2 person-years for 1 year

Research Area: Chemistry/Gas Phase

Activity Title: Evaluate the Role of Clouds in CH_4/CO Tropospheric Chemistry

Description: Evaluate whether clouds play an important enough role in global CH_4/CO chemistry that they should be included in the long-lived species CTM. The vision identified for the long-lived species CTM did not include cloud processes. To a first approximation, this omission is justified because of the relatively small direct impact that aqueous-phase processes have on CH_4 and CO concentrations. However, HCHO, which is an intermediate species in the oxidation of CH_4 to CO, is relatively soluble in cloud and precipitation droplets and its distribution may be significantly affected by cloud processes. An investigation should be carried out to evaluate the potential significance of the neglect of these processes and to recommend alternative designs of the long-lived species CTM, if necessary.

Level I.	Evaluate available literature on the effect of clouds on CH_4/CO chemistry and make recommendations for implementation in the GTCMS.
Level II.	Perform a box-model or column-model analysis to evaluate the effect of clouds on CH_4/CO chemistry and make recommendations for implementation in the long-lived species CTM.
Level III.	Perform a full three-dimensional model analysis to evaluate the effect of clouds on CH_4/CO chemistry and make recommendations for implementation in the long-lived species CTM.

Estimated Level of Effort and Time:	Level I.	0.5 person-year for 1 year
	Level II.	1 person-year for 1 year
	Level III.	2 person-years for 1 year

Research Area:	Chemistry/General
Activity Title:	Develop an Interface Between the Short-Lived Species and the Long-Lived Species and Aerosol CTMs
Description:	Develop the design of the structural interface and methodology to allow the short-lived species CTM to provide oxidant fields to the long-lived species and aerosol CTMs. As described in the research activity "Evaluate Use of the Short-Lived CTM in Providing OH Fields for Use in the Long-Lived and Aerosol CTMs," one way to implement the GTCMS would be to design the long-lived species and aerosol CTMs as engineering models. Full short-lived species CTM simulations would provide oxidant fields for detailed scenario analyses performed by the less computationally expensive engineering models. Once the engineering model concept has been verified, an interface should be designed to allow the oxidant fields to be efficiently transferred from the short-lived species CTM to the engineering models. The interface should allow for both a "screening mode" (with coarse temporal and spatial resolution) transfer of information and a "detailed mode" (with fine temporal and spatial resolution).
Estimated Level of Effort and Time:	2 person-years for 1 year

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Research Area:	Chemistry/General
Activity Title:	Evaluate Use of the Short-Lived CTM in Providing OH Fields for Use in the Long-Lived and Aerosol CTMs
Description:	Evaluate the feasibility of using OH, O_3, and H_2O_2 fields generated by the short-lived species CTM as input data for the long-lived and aerosol CTMs. One way to implement the GTCMS would be to design the long-lived species and aerosol CTMs as engineering models. Full short-lived species CTM simulations would provide oxidant fields for detailed scenario analyses performed by the less computationally expensive engineering models. An evaluation must be performed to establish the feasibility of this design.

Estimated Level of Effort and Time: 1 person-year for 3 years

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Research Area: Chemistry/General

Activity Title: Evaluate Adequacy of Gas-Phase Chemical Oxidation Mechanisms of DMS and the Resulting Production of Cloud Condensation Nuclei

Description: Evaluate the current understanding of the gas-phase chemistry of DMS in the troposphere, including production of SO_2 and methanesulfonic acid (MSA) and the relative amounts produced of each. Examine existing mechanisms and evaluate their predictive ability based on available DMS/SO_2/MSA measurements. Also evaluate the current understanding of cloud condensation nuclei (CCN) production from DMS oxidation products. Develop an appropriate gas-phase chemical mechanism for DMS oxidation for the GTCMS and provide evaluated parameterizations for the production of CCN from DMS oxidation products.

Level I. Compile and evaluate existing mechanisms for DMS oxidation and parameterizations for CCN production and make appropriate choices for the GTCMS, based on current literature.

Level II. Develop a new DMS oxidation mechanism and a new CCN parameterization for the GTCMS, based on the most recent laboratory and field data.

Estimated Level of Effort and Time: Level I. 1 person-year for 1 year

Level II. 2 person-years for 2 years

Research Area: Dynamics/Stratosphere

Activity Title: Develop Stratosphere/Troposphere Exchange Parameterization

Description: Exchange of trace species mass between the stratosphere and troposphere is expected to dominate the upper boundary condition for a tropospheric chemistry model. This exchange must be accurately simulated in the GTCMS. Higher-order resolution modeling and observations should be used to develop and test a parameterization for the exchange of trace species mass between the stratosphere and troposphere suitable for use in a global-scale model.

Estimated Level of Effort and Time:	2 person-years for 3 years

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Research Area:	Dynamics/Boundary Layer Turbulence
Activity Title:	Develop Nonlocal Turbulence Closure Methods for Global Models
Description:	Nonlocal turbulence closure methods allow transport of trace species, heat, and water vapor against the gradient. This is possible because turbulence can transport parcels with high (low) concentrations through layers with low (high) concentrations without mixing. Application of nonlocal turbulence closure to regional-scale models has resulted in significant differences to regional-scale transport. However, the methods used need to be refined to improve the efficiency of computations needed.
Estimated Level of Effort and Time:	1.5 person-years for 3 years

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Research Area:	Surface Exchange
Activity Title:	Synthesize Emissions, Surface Flux, Deposition, and Hydrologic Models
Description:	A single, selfconsistent modeling framework is needed for hydrologic, emissions, surface flux, and deposition models. This framework can then be used to synthesize these models into a single-surface flux module for the GTCMS that has the capability to handle a wide range of surface types, including vegetation, wetted surfaces, water, and land.
Estimated Level of Effort and Time:	2 person-years for 3 years

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Research Area:	Transport/Grid
Activity Title:	Develop an Irregular Grid Method for Chemistry
Description:	Develop the means to apply an irregular grid, or more specifically, a nested grid system to the chemical mechanism. Methods are needed to define the inflow and outflow conditions between the nested and global grid systems in an efficient manner. The method must demonstrate an accuracy within the nested grid that is equal or better than in the global grid.

Estimated Level of Effort and Time: 2 person-years for 2 years

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Research Area: Numerics

Activity Title: Develop a Computationally Fast Chemistry Integrator

Description: A computationally fast and flexible chemistry integrator with the computational complexity of Sillman (1991) and quasi-steady-state approximation (QSSA) (Hesstvedt et al. 1978) is needed to accommodate a wide range of chemical mechanisms and to adapt easily to new mechanisms. The QSSA method handles stiffness by eliminating stiff species and lumping together species of similar chemical type. The code for the QSSA method needs to be modified when the mechanism is changed. The proposed integrator will generate automatically the differential equations, reorder species (as done by Sillman 1991) and use implicit Runga Kutta methods, which are suitable for stiff equations yet have reduced computational complexity because of the reordering of species.

Estimated Level of Effort and Time: 2 person-years for 3 years

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Research Area: Uncertainty

Activity Title: Demonstrate Effectiveness of Autodifferentiation

Description: Techniques have recently been developed to automatically generate a computer code that provides the sensitivity coefficients for a given set of equations that are to be solved numerically. The effectiveness of these techniques, with the equations being solved for tropospheric chemistry, cloud microphysics, and aerosols, needs to be demonstrated. The techniques should be applied to a module for which sensitivity coefficients can be determined by other means, so that the results can be compared and the effectiveness of the method evaluated.

Estimated Level of Effort and Time: 1 person-year for 1 year

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Hesstvedt, E., O. Hov, and I.S.A. Isaksen. 1978. "Quasi-Steady-State Approximations in Air Pollution Modeling: Comparison of Two Numerical Schemes for Oxidant Prediction." Int. J. Chem. Kinetics X:971-994.

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