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PRECIPITATION SCAVENGING MODELS: SENSITIVITIES, TESTS, AND APPLICATIONS

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PRECIPITATION SCAVENGING MODELS: HISTORY AND FUTURE DIRECTIONS

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ABSTRACT

"Multiphase" atmospheric-chemistry models can be described as atmospheric-pollutant simulations that explicitly differentiate between physical phases in the atmosphere (e.g., gas, cloud water, rain water, snow, · · ·), and directly compute chemical transport and transformation behavior between and within each of these individual phases. Initially formulated for specific application to precipitation-scavenging analysis, many attributes of these models have become incorporated into the more general atmospheric-chemistry codes as well.

During the past few years, several of these multiphase precipitation-scavenging models have been developed to the point where they can be applied, in a moderately straightforward fashion, by members of the extended atmospheric sciences community. This presentation provides a brief overview of several aspects of a number of these models, including their structure, their application, their sensitivities and uncertainty levels, their evaluation against field measurements, and their availability.

1. INTRODUCTION

Our evolving understanding of wet-removal processes and precipitation chemistry can be generally characterized in terms of two broad and overlapping eras. The first of these, which we shall refer to as the **consolidated parameter era**, began essentially with Chamberlain's pioneering work in the early 1950s (Chamberlain 1953). As its name implies, this era involved attempts to quantify the relationship between airborne pollutant concentrations, meteorological conditions, and wet-delivery rates by lumping the effects of multiple attachment and delivery mechanisms in terms of single, consolidated parameters.

Chamberlain's early work introduced the concept of the *scavenging coefficient* as a consolidated parameter. Although first applied to characterize relatively simple situations involving below-cloud scavenging of aerosol particles and soluble gases by rain, this

parameter was soon extended in attempts to describe total storm situations of varying degrees of complexity (e.g., Lang and Knox 1977). The scavenging-coefficient concept was eventually followed by similar applications of other consolidated parameters, such as the *in-cloud scavenging efficiency* (Junge 1963) and the *scavenging ratio* (Engelmann 1971). Figure 1 presents a general historical overview of some of these concepts, in the context of several major "issues" to which they have been applied.

It is important to note that these types of consolidated parameters have substantial basis in theory, at least as applied to relatively ideal meteorological situations. Regardless of these theoretical underpinnings, however, the consolidated-parameter approach has experienced two major shortcomings. First, there has been a substantial tendency to extend the use of consolidated parameters to characterize situations far less ideal and more complex than originally envisioned. In most such situations the parameters lose much of their interpretive significance and move strongly in the direction of total empiricism. Second, the lumped characterization of multiple mechanisms has the disadvantage of obscuring the effects of individual rate-influencing phenomena. This generally results in an unacceptably wide scatter in measured values of these parameters, and a corresponding uncertainty in their practical application. Moreover, the direct mechanistic interpretation of complex scavenging processes is usually precluded, or at least severely clouded, with this approach.

The second broad era of understanding is characterized by attempts to express individual mechanistic phenomena explicitly, rather in terms of lumped parameterizations, and shall be referred to here as the **mechanistic era**. As with its consolidated-parameter counterpart, this era began with relatively simple applications to below-cloud systems (e.g., Drewes and Hales 1982), but was soon extended to more complex systems involving total storms. Quite obviously, efforts in the second era tend to be computationally intensive and they have become possible only with the advent of modern computers.

The temporal overlap between these two eras is considerable. Although the accomplishments to date in the mechanistic era have been substantial, a large effort continues to be applied using consolidated parameters. Two reasons for this are apparent. First, consolidated parameters have the distinct advantage of simplicity, thus making them convenient for expressing observational data and applying them in terms of simple assessment models. Second, the computer codes associated with the second era tend to be localized--usually at the institutions at which they have been created--and thus are not considered generally available for use by the scientific community.

The basic theme of the present paper is the demonstration that these mechanistically oriented computational tools are indeed becoming generally available for widespread use, suggesting that they should be applied in an increasingly routine fashion in the future. This will be accomplished by providing a brief overview of contributions to the mechanistic era, from the perspective of the scientist who desires to interpret observed wet-removal behavior on a detailed mechanistic basis.

2. MULTIPHASE ATMOSPHERIC CHEMISTRY MODELS

Probably the most significant contributions within the mechanistic era to date have been associated with the development of multiphase atmospheric chemistry models. These models take the form of atmospheric-pollutant simulation codes, which explicitly differentiate between physical phases in the atmosphere (e.g., gas, cloud water, rain water, snow, $\cdots$), and directly compute pollutant transport and transformation behavior between and within each of these physical phases. Such codes essentially provide a computational forum that allows explicit characterization of cloud-physics processes, pollutant-attachment phenomena, chemical reaction, and additional steps of the scavenging sequence, in a mathematically accountable and physically realistic fashion.

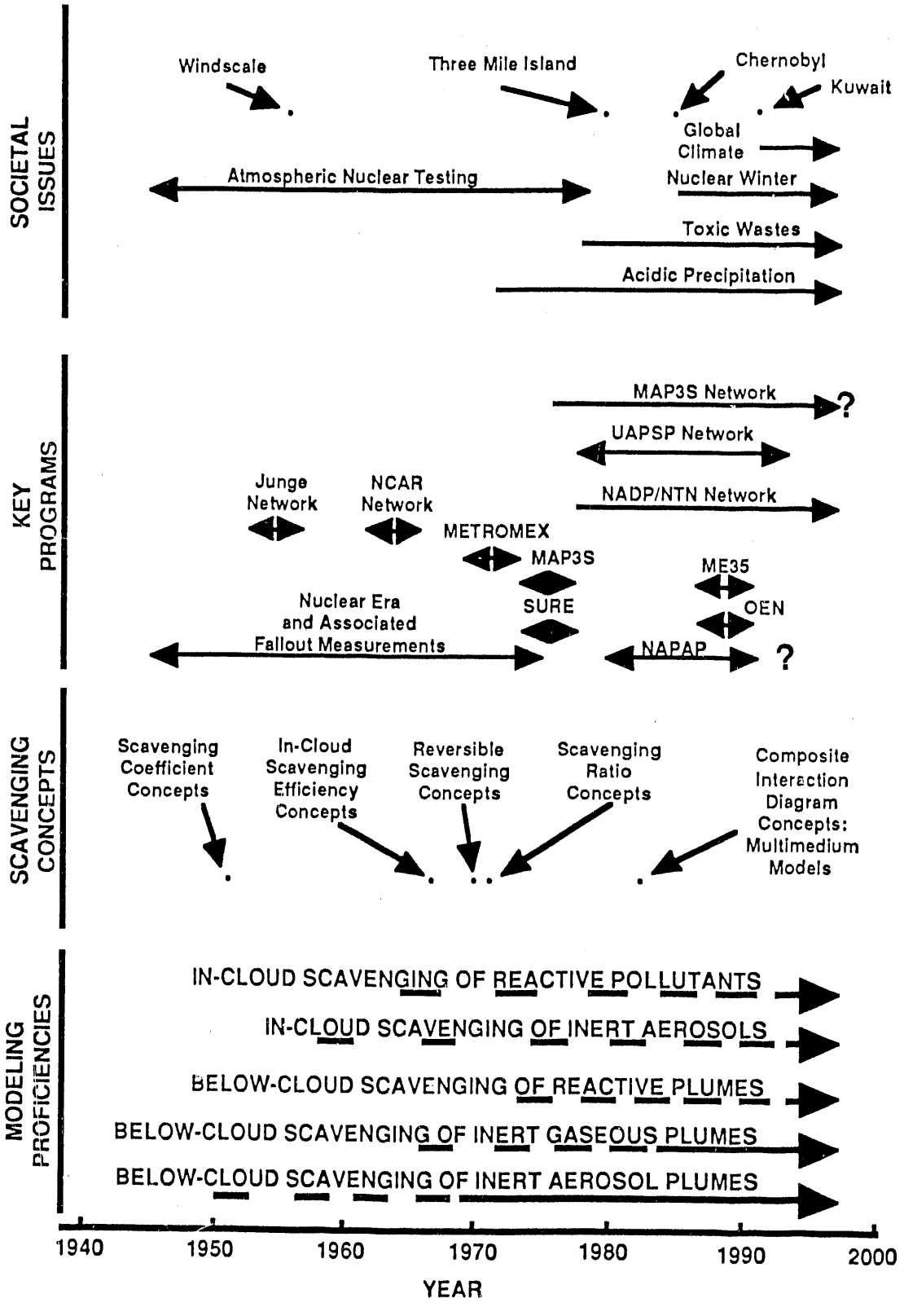

FIGURE 1. Historical Chart for Scavenging-Concept Development

Figure 2 presents a qualitative idea of how these processes are incorporated within a given code. This indicates three coupled "interaction diagrams," corresponding to water, sulfur pollutants, and nitrogen pollutants, which have been implemented within a code execution. Here the arrows between the various elements have been incorporated into the code in the form of mathematical rate expressions corresponding to the individual mechanisms. Typically the code is written in a modular fashion, so that modifications to any given mechanistic scheme can be incorporated simply by recoding the individual rate expressions.

Some examples of mechanistic model output are given in Figure 3, which shows output from a two-dimensional simulation of a warm-frontal storm. While these plots will not be discussed further here, the reader can easily recognize the high degree of detail provided by such simulations and their potential for in-depth diagnostic analysis of mechanistic behavior.

Table 1 summarizes some of the existing multiphase reactive storm codes that have been created to date. While this listing is confined primarily to mesoscale codes designed for the scientific interpretation of wet-removal behavior, it should be noted that the more modern regional and global assessment codes (e.g., Carmichael *et al.* 1991, Berge 1990, Chang *et al.* 1987, Luecken *et al.* 1991) are beginning to adopt some of the elements of this technology. These mechanistic features can be expected to migrate to the large-scale models even more extensively in the future, as computer speeds increase.

3. MODEL SENSITIVITY ANALYSES AND MODEL EVALUATIONS

The large number of species and physicochemical mechanisms typically associated with multiphase scavenging models presents a major problem in their application for interpretive analysis. In essence these variables correspond to multiple degrees of freedom, and it is often difficult to determine, simply from comparison of simulations with observed data, which mechanisms are responsible for specific features of interest. This problem carries over into the issue of model evaluation as well. Typically it is extremely difficult to perform model-evaluation field studies that successfully measure all of the physical and chemical features that are necessary for a totally unambiguous test of model proficiency and credibility.

These problems have resulted in an increasing level of interest in the *sensitivity analysis* of scavenging models, wherein the quantitative dependence of model output on individual mechanisms and/or model inputs is evaluated. A number of published studies (e.g., Hegg and Larson 1990, Tsai and Altwicker 1990, Easter and Luecken 1988) have approached this issue in a straightforward manner, simply by conducting repeated code executions while varying selected parameters among the individual executions, and observing the associated code outputs. A significant further effort, however, is currently developing in the application of more sophisticated and efficient mathematical techniques for detailed local and global sensitivity analyses (e.g., McCroskey and McRae 1987, Kramer *et al.* 1982, Cho, Carmichael, and Rabitz 1987, 1988), and formally extending these concepts for the objective estimation of parameters (McCroskey 1988). While much of the theoretical development in these areas is associated with other applications such as chemical-engineering process design, these methods can be expected to receive extended application in scavenging models during the next several years.

Although no truly comprehensive set of field data is currently available for definitive scavenging-model evaluation, several limited field studies have generated data having the potential of providing considerable insight into model proficiency. Probably the most well-known of these is the so-called OSCAR study (Chapman, *et. al.* 1987) which resulted in a meteorological and chemical data set for springtime storms in the midwesteren U.S. A number of subsequent studies, notably those in the DOE PRECP series (cf. PRECP, 1986) have attempted to characterize the dynamical and chemical nature of various types of North

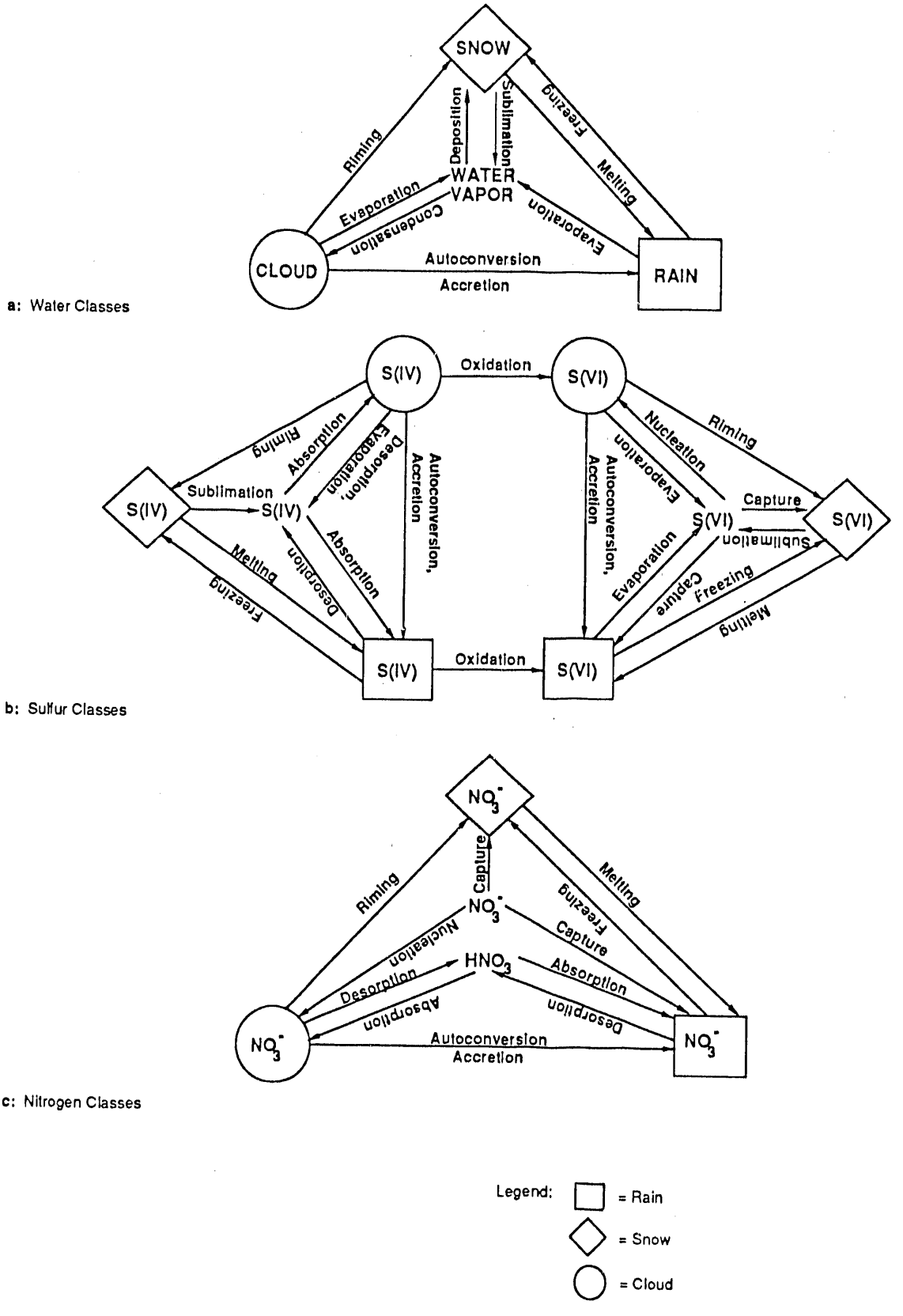

FIGURE 2. Example Interaction Diagrams

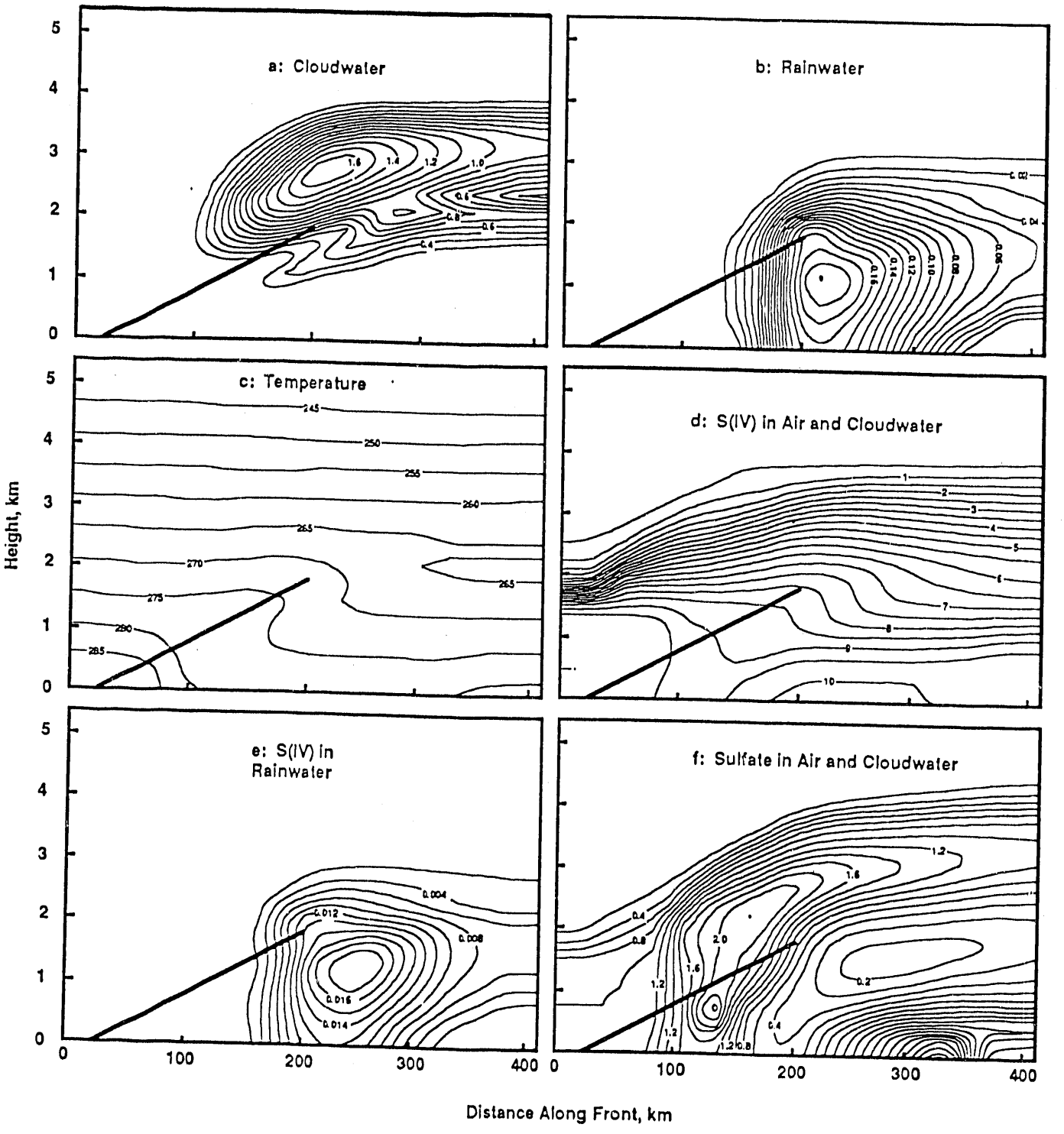

FIGURE 3. Example Output From a Two-Dimensional Reactive Storm Model. Contour Levels for Sulfur Compounds Denote Mixing Ratios in Nanomoles per Mole Air (ppb). Levels for Water Species Denote Mixing Ratios in Milimoles per Mole Air. Temperature is in Degrees K. Diagonal Line Depicts Position of Frontal Surface. Warm-Front Storm Simulation From Hales (1989).

Table 1: Summary of some mesoscale mechanistic scavenging models

Code Type (or Name)	Dimensionality	Descriptive Summary	References
PLUVIUS (General)	1,t or 2 ss	Kinematic; i.e., based on material and energy balances only. thus requiring wind-field input from external sources. Generalized and modular; intended for widespread, routine application	Hales (1982) Easter and Hales (1984) Easter and Hales (1983)
Convective Storm	3,t	Kinematic; dynamic driver	Tremblay and Leighton (1986) Tremblay (1987)
Warm-Frontal Rainband	2,ss	Kinematic	Hegg et al. (1984)
General	1,ss	Kinematic	Tremblay and Leighton (1984)
Cold-Frontal Rainbands	2,ss	Kinematic	Rutledge et al. (1986) Hegg et al. (1986)
Convective Storm	1.5,t	Dynamic; i.e., based on momentum equation as well as material and energy balances. Generates wind fields internally	Lee (1986)
Convective Storm	1,t	Kinematic	Niewiadomski et al. (1986)
Frontal Storm	1,t	Kinematic	Kavassalis et al. (1986)
Frontal Rainbands; Adaptation of Tremblay-Leighton (1986) Model	2,t	Kinematic; dynamic driver	Leighton et al. (1990)
Meso-STEM (General)	2,t and 3,t	Kinematic; dynamic driver, highly modularized and flexible chemistry and physics	Hong and Carmichael (1986) Shim and Carmichael (1991)
Orographic and Frontal Systems	3,t	Kinematic; dynamic driver, two-parameter descriptions (log-normal) of cloud droplet, raindrop and aerosol size-distributions.	Chaumerliac et al. (1987) Chaumerliac et al. (1990)
Stipulated Rain Rate	1,t	Kinematic; very simple microphysics	Koo et al. (1990)
Convective Storm; PLUVIUS Adaptation	1,t	Kinematic; applied to specific observed storms	Tsai et al. (1990)

Table 1, Continued

Convective Storm; PLUVIUS Adaptation	1,t	Kinematic; applied to specific observed storms	Tsai et al. (1990)
PLUVIUS II	1,t, 2,t, or 3,t	Kinematic; generalized and modular; intended for widespread, routine application	Easter and Luecken (1988) Hales (1989) Hales and Easter (1991)
RSM	1,t	Kinematic; model adapted from a scavenging module based on PLUVIUS II; generalized and modular; prescribed microphysics	Berkowitz et al. (1989)
Maritime Warm-Rain Cloud	2,t	Kinematic; dynamic driver; detailed descriptions of cloud droplet, raindrop, and aerosol size distributions	Flossmann and Pruppacher (1988)
Horizontally Homogene-ous Storm	1,t	Kinematic; prescribed wind fields	Molenkamp (1983)

American storms, including thunderstorms, northeastern snowstorms, and mid-latitude cyclones. A more recent field effort, known as the DOE Frontal Boundary Study (FBS), has probably resulted in the most comprehensive data base to date. Conducted during the late fall of 1989, this study focussed on frontal storms in the upper midwest and obtained high-resolution measurements of meteorology and precipitation chemistry, with rather comprehensive aircraft observations as well. Data processing and reporting for the FBS is currently in progress.

More comprehensive measurement sets are definitely required for definitive model evaluation. The data from these completed studies is underutilized at present, however, and significant new information regarding model performance and mechanistic behavior is expected to result from their future analysis in concert with the refined sensitivity evaluations described above. Much remains to be done in this important research area.

4. CONCLUSIONS

As a concluding point to this paper, it should be reemphasized that the so-called "mechanistic" scavenging codes have developed to a point where their use is common and that all scientists dealing with precipitation-scavenging analysis should consider their application to specific problems at-hand. Although many of these codes are essentially "in-house" programs, a growing number (e.g., the PLUVIUS series) have been documented in user's manuals and are intended for widespread application. Such application, especially in conjunction with advanced sensitivity and parameter-estimation procedures should see extensive application in the future.

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DISCUSSION

C. J. WALCEK. Current "scavenging" models of cloud systems tend to emphasize only one or two of three areas of scientific focus: dynamics (e.g., Clark); chemistry (e.g., Jacob, Graedel); or microphysics (e.g., Hegg, Charlson). What is your impression of the balance of these three scientific study areas in existing models, and how will this change in the future?

J. M. HALES. I agree with you that any given one of today's scavenging models tends to emphasize one or two aspects of the total picture at the expense of others. This, I feel, results from two basic causes: one, the tendency for a disciplinary approach to a problem that is truly multidisciplinary in scope and two, the past lack of computer resources to deal with the problem in its totality. I strongly suspect that both of these constraints will diminish in the future and that you will begin to see a much more *gestalt-oriented* approach taken during future years.

L. A. BARRIE. Scavenging models are based on the solution of a nonlinear differential equation set. Chaos theory has begun to show us that in the solution space of such sets there exist domains where unique solutions are not possible. To what extent in the current models are chaotic regimes allowed, and if so do you see evidence of them?

J. M. HALES. Chaotic behavior, if it proves to exist in these situations, will arise naturally in the solutions of the model equations. I can't give you a definitive answer as to how important this may be in all situations, but I can give some insights from my experiences in generating PLUVIUS II results, such as those shown in Figure 3. I produced these plots by initializing the model and then executing for prolonged lengths of time in an attempt to approach some sort of steady-state conditions; thus the storm cross sections that appear here correspond to snapshots in time which -- if a true steady-state exists -- should not vary from one time to the next. Indeed they <u>did</u> vary somewhat, exhibiting limit cycles that shifted the results periodically with time. The magnitude of these cycles was probably not more than five to ten percent of the mean values, however; thus chaos seems to be of minor concern in this case.

S. E. SCHWARTZ. Following Dr. Barrie's question, I understand that for a convectively unstable atmosphere the initiation of a storm is very susceptible to minor influences. How do you initiate storms in your model?

J. M. HALES. Remember that PLUVIUS II is a kinematic code; in other words, it does not solve a momentum balance and thus requires wind fields to be supplied from external sources (e.g., output from a dynamic model). As such PLUVIUS II initiates storms simply as the consequence of feeding water vapor into the pre-defined flow system, and doesn't have to worry much about the minor influences that you mention. These are important, however, to the dynamic code that generates the winds. Typically these codes are initialized with an unstable atmosphere and start the storm simply by perturbing the numerical system at some point in space and time.

V. M. VOLOSHCHUK. What is the air volume, according to the model calculations, which cleans, on average, a cloud formation during its lifetime (in units of its volume)?

J. M. HALES. This obviously depends on the lifetime of the cloud. The frontal storm shown in Figure 3 (a quasisteady-state) has an indefinite lifetime and thus the amount of air processed will be very large (in this case it takes about ten hours for an air parcel, travelling at 20 m/sec, to enter the system from the left and exit at the right). For more transient clouds this volume will be less. I expect, for example, that some convective storms process about ten cloud volumes of air before dissipating.

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