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Uncertainty in Modeling Regional Environmental Systems: The Generalization of a Watershed Acidification Model for Predicting Broad Scale Effects

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IIASA Research Report
April 1990

UNCERTAINTY IN MODELING REGIONAL ENVIRONMENTAL SYSTEMS:
The generalization of a watershed acidifcation model for predicting broad scale effects

Jean-Paul Hettelingh

RR–90–3
April 1990

Reprint of PhD Thesis submitted to the
Free University, Amsterdam, 1989

INTERNATIONAL INSTITUTE FOR APPLIED SYSTEMS ANALYSIS
Laxenburg, Austria

Reprint of PhD Thesis submitted to the Free University, Amsterdam, 1989.

Printed by Novographic, Vienna, Austria

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FOREWORD

Acidification of bodies of freshwater was one of the first effects of transboundary air pollution to be noticed in Europe. The Regional Acidification Information and Simulation (RAINS) model developed within the Acid Rain Project and later the Transboundary Air Pollution Project at IIASA contains a Lake Acidification submodel. Jean-Paul Hettelingh was a key member of both projects in developing that submodel, while working on his Doctorate at the Free University of Amsterdam. This Research Report, in fact Dr. Hettelingh's thesis, describes the development of the Lake Acidification submodel of RAINS and its application to whole regions, and in particular the Netherlands and Norway. A unique feature of this work is the concept of "flexible zoning" in which model predictions are obtained as a function of model calibrations within predetermined regions.

R.W. SHAW, *Leader*
Transboundary Air Pollution Project

B.R. DÖÖS, *Leader*
Environment Program

ABSTRACT

Policies aimed at the alleviation of negative environmental impacts have increasingly been based on predictions made with models of the environmental systems involved. The usefulness of these models is limited by many uncertainties, e.g., is the complexity of a system properly reflected in the model structure and chosen aggregation-level? Is the data used in the model representative of the system? Is the temporal and spatial scale used in the model appropriate for understanding the system's behavior? The combination of these uncertainties may lead to unexpected results.

This study identifies key factors determining watershed responses to acid deposition in different regions, using the RAINS Lake Acidification Model. The study aims at:

- Providing a method for defining a region such that environmental policy directed at alleviating watershed acidification will become more suitable for the large scale management of surface water quality.
- Depicting regional characteristics that allow for the usage of a less detailed and thus more aggregated model.
- Providing an operational concept of critical loads for policy insight into watershed quality as a function of the spatial zoning of watersheds.

The results of the study are:

- The 5-step method of flexible zoning introduced in this study, allows for a probabilistic investigation of the compatibility between models and available spatial data.
- The calibration of models to previously defined regions may be of limited use for policy purposes because predictions of environmental effects (i.e., watershed acidification) as a result of changing deposition patterns over large regions may be error prone.
- Cumulative distributions of model predictions about the acidification of watersheds should be used to assess critical and target loads for broad regions.

Key words: acidification, aggregation, calibration, critical loads, environmental modeling, sensitivity analysis, uncertainty analysis, regionalization, target loads.

A la mémoire de mes grands-parents,
Angèle et Victor Mennetrier-Galissot

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PREFACE

The major portion of the research for this study was done at the International Institute for Applied Systems Analysis (IIASA) in Laxenburg (Austria). Several important tasks were undertaken at the Oak Ridge National Laboratory (ORNL) in Oak Ridge, Tennessee (USA) during visits there in 1987 and 1988. It gives me great pleasure to thank all those that directly or indirectly contributed to this result.

I would like to express my gratitude to Prof. Dr. Peter Nijkamp of the faculty of economics and Prof. Dr. Hans B. Opschoor of the Institute for Environmental Studies, both at the Free University in Amsterdam (The Netherlands), for their stimulation, comments and criticisms in the course of this study.

This study would not have been initiated without the influence of Dr. Robert H. Gardner's work in the field of uncertainty analysis on environmental models at the Environmental Sciences Division of ORNL. It is therefore difficult to find the proper words for expressing my appreciation to Robert Gardner for his stimulating ideas which supported the study.

The leader of IIASA's Acid Rain Project (ACI), Drs. Leen Hordijk is acknowledged for stimulating, guiding and critically examining the early phases of this study.

Dr. Rod Shaw, the current leader of the Transboundary Air Pollution project (TAP, formerly ACI), is gratefully acknowledged for arranging fiscal support for writing this dissertation. Furthermore I would like to thank him for his inspiring comments on early versions of the manuscript.

Comments on early drafts of the manuscript by Dr. Floor M. Brouwer, my colleague at IIASA and former colleague at the Institute for Environmental Studies at the Free University in Amsterdam, are particularly appreciated.

The library support of Eduard Loeser, Ingrid Guerke and Helga Lowe (IIASA) and of Maud Blaauw (Free University) was of considerable help and assistance of Eva Delpos and Anka James (IIASA) in preparing graphs was indispensable.

My home institute, the Institute for Environmental Studies at the Free University, is acknowledged for providing overhead support in the final stage of this study.

My current employer, the National Institute for Public Health and

Environmental Protection (RIVM) in Bilthoven (The Netherlands) is credited for generously providing me with the time I needed in the last phase.

Current and former colleagues of IIASA's Transboundary Air Pollution project, Joseph Alcamo, Marcus Amann, Stuart Batterman, Jerzy Bartnicki, Pekka Kauppi, Lea Kauppi, Juha Kämäri, Gabor Kornai, Barbara Luebkert, Annikki Mäkelä, and Wolfgang Schoepp provided a warm yet professional atmosphere to get the present study done. In this respect special credit should go to my colleague and room mate, Maximilian Posch, who provided advice, technical assistance and an excellent sense of humor. I am grateful to my friends and colleagues at ORNL, Steve Bartell, Antoinette Brenkert, Bob Cook and Kenny Rose, and to Bob and Sandi Gardner and their children Nathan and Jeremy for their hospitality during the stay of my family and me at Oak Ridge in 1988.

No matter how excellent my professional atmosphere may be, nothing can replace a supporting personal surrounding. My spouse Elisabeth and our daughter, Charlotte, have provided this surrounding in spite of the stresses to the family due to this research. Their influence as well as the warm involvement of my parents Jeannine and Wim were of vital importance to the success of this study. I am also indepted to my brother Frank, his spouse Annette and to my parents-in-law Fernand and Jacqueline Garnier.

The contents, the research design, and the execution of the present study have been independently performed by the author. Any errors, therefore, are my responsibility. This caveat also applies to the typing of the manuscript which was accomplished with WordPerfect 4.2 and printed on a HP laserjet II.

Mödling, May 1989

1. GENERAL INTRODUCTION

1.1 INTRODUCTION

Prologue

No part of the universe is so simple that it can be grasped and controlled without abstraction (Rosenblueth and Wiener, 1945). However, a certain methodological procedure used by investigators to make an abstraction of a part of the universe heavily depends on their perception of the systems that are part of the universe. Some scientists view systems holistically as everything being indisruptably related to everything else. Others adopt the reductionist approach where systems can be understood by mechanistic principles (Hofstadter, 1980, p.389) which explain the whole system on the basis of a detailed understanding of its parts.

The understanding of systems as a collection of parts evolved with the development of industrial organization in the western world. The mechanization of the production and the rapid growth of industrial enterprises required the coordination of many different managerial functions and tasks. The scientific management of many different activities to support a task became known as Operations Research during the second World War. Operations Research provides (industrial) managers with a rigorous basis for solving problems involving the interaction of components of the organization in the best interest of the organization as a whole (Churchman et al., 1957, p.6).

In later years C. West Churchman wrote: "....the earliest document aiming at a systems approach to decision making was written in China in the second millennium B.C.. This very early attempt- which, interestingly enough, became quite popular in the 'radical culture' of the 1970s- is the I Ching or Book of Changes" (Churchman, 1979, p.32). In I Ching the universe is assumed to be holistically organized, such that guidance through daily life may be achieved by consulting randomly obtained signs, called hexagrams. These hexagrams were

intended to serve as a mapping of the universal space of answers to particular problems. Nowadays, consulting such an oracle is far from the way in which policy makers tackle real life problems. On the contrary, policy making consists often of a more mechanistic approach that partitions real life problems into areas of responsibility governed by particular ministries, hierarchical administrations and procedures.

In modern scientific research a similar approach of making abstractions of parts of the universe by mental or formal models, still prevails. The result is that an increasing number of scientific studies of real life processes by means of systems analysis can be found. According to Quade and Miser (1985, p. 2) "the central purpose of systems analysis is to help public and private decision makers to ameliorate the problems and manage the policy issues that they face." Policy making on social, economic and environmental issues must rely on abstract representations of real world systems that can then be simulated by computer modeling, analyzed by mathematical methods and verified by statistical analysis. Although simplicity seems to prevail as an objective of systems analysis, the tools, terminology and interpretation of results can vary greatly among scientific disciplines. The methods used vary from simple data analysis to the use of mathematical equations and solution techniques by means of advanced computer algorithms. These methods have become increasingly tailor-made tools for coherent policy analysis.

A bird's-eye view of the study.

The purpose of this study is to provide policy makers[1] with a comprehensive means of dealing with the definition, localization and assessment of damage to the natural environment from human activities. With the growth of the scale of the interactions between human development and the environment, the complexity of these interactions has also increased. Today's concern about the greenhouse effect and its environmental consequences is an example of an emerging consciousness that human activities, biochemical processes and climate are

[1] Some terminological classification is in order first. In general 'policy makers' are responsible for managerial actions, whilst 'policy analysts' provide the technical support to a 'policy maker'.

interrelated (see also Clark and Munn, 1986). Individual environmental problems like acid rain, soil erosion, ozone depletion, deforestation and surface water pollution are all associated with interrelated human activities. Studying these problems requires a multi-disciplinary scientific collaboration and an integrative - or at least coherent- way of describing and formalizing the system of causes and effects. Disentangling relationships between subsystems implies observation and analysis on more than one level, as is illustrated by the question whether climatic change is due to the depletion of ozone or to chemical processes between hydrocarbons and nitrate-oxides which indirectly affect the ozone concentration. These questions often fall into the category 'what was first; the chicken or the egg', and may need a hierarchical arrangement of complex systems to efficiently address these issues (Mesarovic, 1984). Investigating global change of the environment requires an understanding of the hierarchy of causes and environmental problems as well as an identification of their related temporal and spatial scales. The temporal scale of environmental problems often results from economic and social activities that are often undertaken for a benefit that is measurable within the time span of the current human generation. The usage of pesticides, for example, may have increased the quantity of agricultural output over the past years but the persistence of these pollutants may jeopardize the quality of natural products for a long time to come. The related spatial scale may extend far beyond the regional scale within which the economic and social development occurred. Many studies of interactions between activities and environment concentrate on individual rather than on global problems, on immediate impacts of ameliorative measures rather than on a sufficiently long time horizon, and on local or national scales rather than on a sufficiently broad spatial scale.

However, this study had to be kept to manageable proportions. It will indirectly deal with general issues like 'hierarchical arrangements' and 'complexity' of systems only to a limited extent. The central question here can be formulated as: what level of detail is necessary to be able to predict effects over the broad spatial scales relevant for policy design and policy measures? The acidification process, for example, of one single lake in a given area can be reasonably understood and modeled. The questions to be addressed here are (1) to what extent and level of confidence can this model be used

to predict the acidification of many lakes over a broad spatial scale? (2) how should the region be defined to reflect the model's input needs and output capabilities?

The problem of defining the scale at which model application is justified has some similarity to the modifiable areal unit problem. The modifiable areal unit problem is used in the spatial planning sciences to describe the uncertainty concerning the identification of objects in a spatial study (see also Openshaw, 1978). In the discipline concerned with spatial behavior problems, data are available that have been spatially aggregated more than once. These data then serve as input values for spatial interaction models leading to model predictions that may bear behavioral significance but, on the other hand, might very well only reflect the way in which a study region is partitioned into zones (Openshaw, 1977b). Similarly, the prediction of economic models that describe the development of economic variables, e.g. industrial production and investments, on a macro economic scale is only equal to the sum of predictions obtained by applying these models on various micro economic scales if particular conditions are fulfilled (see also van Daal and Merkies, 1984). Regional economic models are often tailored to the regional scale at which data is available. Spatial units are often not appropriate for the usage of economic models for regional economic policy analysis. Therefore, emphasis is often put on statistical techniques to make optimal use of available temporal and spatial data (see also Folmer, 1986). Economic functions or prevailing administrative structures may have influenced the composition of these regional scales (Nijkamp et al., 1984). The categorization of social and economic functional relationships in the Netherlands, for example, has lead to the distinction of 40 COROP regions (CBS, 1975) for which data is available that is used by a great variety of models (see for example Arntzen et. al., 1981; de Vries, 1987).

Thus the questions formulated above have some relevance for many investigations that involve the application of models on different spatial scales. In this study the choice was made to restrict these investigations to the environmental system.

This study does not deal with spatial interaction patterns and related models in general, but deals more specifically with the relationship between zoning of an environmental system and its consequences for environmental model results.

The environmental process chosen in this study is watershed acidification on scales that vary from a single lake to large regions containing many lakes. The study is a technical exercise that needs to be treated in a broader frame because of the relation between system complexity, different aggregation levels, model simplification, zoning and model prediction uncertainty.

The precise aim and restrictions of this study are described in section 1.4. Since, in the disciplines dealing with systems representation a unified terminology is lacking, the concept of systems and models in general and the development of model building and methodological applications in particular will be reviewed first in this chapter. Then environmental modeling is treated in terms of policy use, real life system interactions, model structure and model aggregation (i.e., macro versus micro) in relation to model result confidence and methods to perform uncertainty analysis (Chapter 2). These theoretical sections are followed by a description of the subsystems represented in the so called RAINS Lake Model[2], used in this study, and the equations describing the chemical processes that lead to the acidification of a single watershed (Chapter 3).

The RAINS Lake Model is then used to evaluate a method, commonly referred to as calibration, for matching model results to zonal (regional) data (Chapter 4). Finally a new method, flexible zoning, is proposed in which regional model results, are evaluated in conjunction with uncertainty analysis of model variables that are important in each zone (Chapter 5). Chapter 6 then discusses the application of flexible zoning of the RAINS Lake Model in the Netherlands. A retrospective and prospective chapter concludes this study.

1.2 SYSTEMS AND MODELS

Terminology and concepts of systems

Following Fishman (1973), Kleijnen (1974) and Rubinstein (1981) a

[2]The Regional Acidification INformation and Simulation model describes causes and effects of acidification in Europe. It consists of several modules (see Chapter 2) including the simulation of lake acidification.

system is considered to consist of a set of related components or elements, each having certain characteristics or attributes with numerical or logical values. System elements may be related to one another by intra-relationships, whereas relationships with elements of another, usually external, system are defined as inter-relationships. The set of external systems is called the systems environment. The systems environment generates inputs that are transformed by the system to become outputs. The transformation of inputs is generated by the process operating the system.

Systems may show varying values of its attributes over time. Such dynamic behavior is denoted by the state of a system. If the probability of the system in some specific state does not vary over time, the system is said to be in steady state. Otherwise its state will be called transient.

Systems may be classified in many ways. Kleijnen (1974, p.3) for example, mentions real world vs abstract, black box vs identified, open vs closed, adaptive vs nonadaptive, feedback, feedforward and non-controlled, static vs dynamic, stochastic vs deterministic, continuous discrete and hybrid, technical, organizational and abstract systems. Each discipline tends to have its own systems approach (see for example Emery, 1969; Lasker (ed), 1981). Odum (1986) for example defines an ecosystem as the sum of the input environment, the area with its biota and the output environment.

In all disciplines nowadays an effort is made to recognize a system and its inputs, outputs and state to be able to make predictions, i.e. to estimate the value of a systems output when its inputs change, or to evaluate a system's state when some of its attributes reach other (perhaps critical) values. To understand a system, its hierarchy and its different levels of detail need to be investigated in order to adequately describe the model's operations either verbally, mathematically or technically. Any of these descriptions of a system can be termed a model.

Terminology and model concepts

The aims of using a model are, according to Fishman (1973, p.11):

1. enable an investigator to organize his theoretical assertions

and empirical observations about a system and to deduce the logical implications of this organization,

2. lead to improved system understanding,
3. bring into perspective the need for detail and relevance,
4. expedite the speed with which an analysis can be accomplished,
5. provide a framework for testing the desirability of system modification,
6. make easier and less costly to investigate system's behavior, and
7. permit control over more sources of variation than the direct study of a system would allow.

Models, like systems, have in the past been classified by many authors in different ways (see also Mihram 1972, pp.3-11). In the early days of systems representation one was concerned with the definition of different kinds of models. Rosenblueth and Wiener (1945) distinguished between (1) Material models, which consider transformations of original physical objects, and (2) Formal models, which provide a symbolic, mathematical representation of the original factual system. These formal models were sub-categorized in (1) open-box models which were predictive models for which, given all inputs, the model output could be determined and (2) closed-box models, which were explorative models of a more descriptive nature aimed at understanding a system's output under differing input conditions. With the increasing availability of computing machines, algorithms for symbolic models "were no longer constrained to provide the time-stepped solution of a few differential equations, but could be employed to simulate the behavior of large numbers of elements in complex, interactive systems" (Mihram, 1974, p.5). The use of computerized symbolic models became a primary tool of the systems analyst, which led to the distinction of iconic, analog and symbolic models (Churchman et al., 1957). Iconic models pictorially represent certain aspects of models; analog models employ one set of properties to represent some other set of properties that the system under study possesses; and symbolic models require mathematical or logical operations that can be used to formulate a solution to the problem at hand. Symbolic or formal models were used to describe the same kind of system's representation that could be sub-categorized into

analytic and numerical models. However, this distinction did not lead to an operational difference; both kind of models consisted of a set of mathematical equations of which only the solving techniques were supposed to be different.

The early emphasis on system description, model definition and model solving has gradually been replaced by an extension of the aim of modeling, viz. from solving, towards simulation.

The simulation of systems

This section will present an overview of the terminology used by different authors in an attempt to distinguish the different kinds of simulation and their application. A summarizing synthesis of the terminology and types of model application distinguished by the different authors treated below, is presented in Figure 1.1.

The new modeling capability of simulation was incorporated by Sayre and Crosson (1963), who introduced three categories of models:

1. replication: models that display a significant degree of physical similarity between the model and its representation of the system (e.g. a flight simulator).
2. formalization: symbolic models in which none of the physical characteristics of the modeled system is reproduced in the model itself, and in which the symbols are manipulated by means of an exact discipline such as mathematics or logic, and
3. simulations: symbolic models in which none of the physical characteristics of the model is reproduced in the model itself, and in which the symbols are not manipulated entirely by an exact discipline in order to arrive either at a particular numerical or at an analytic solution.

Analogy exists between some of the model definitions introduced above. Formalized models are similar to the formal models in the terminology of Rosenblueth and Wiener's (1945), and to the symbolic models of Churchman et al. (1957). The equations of formalized models became tractable by means of algebra and calculus, which is not always true for the class of simulation models.

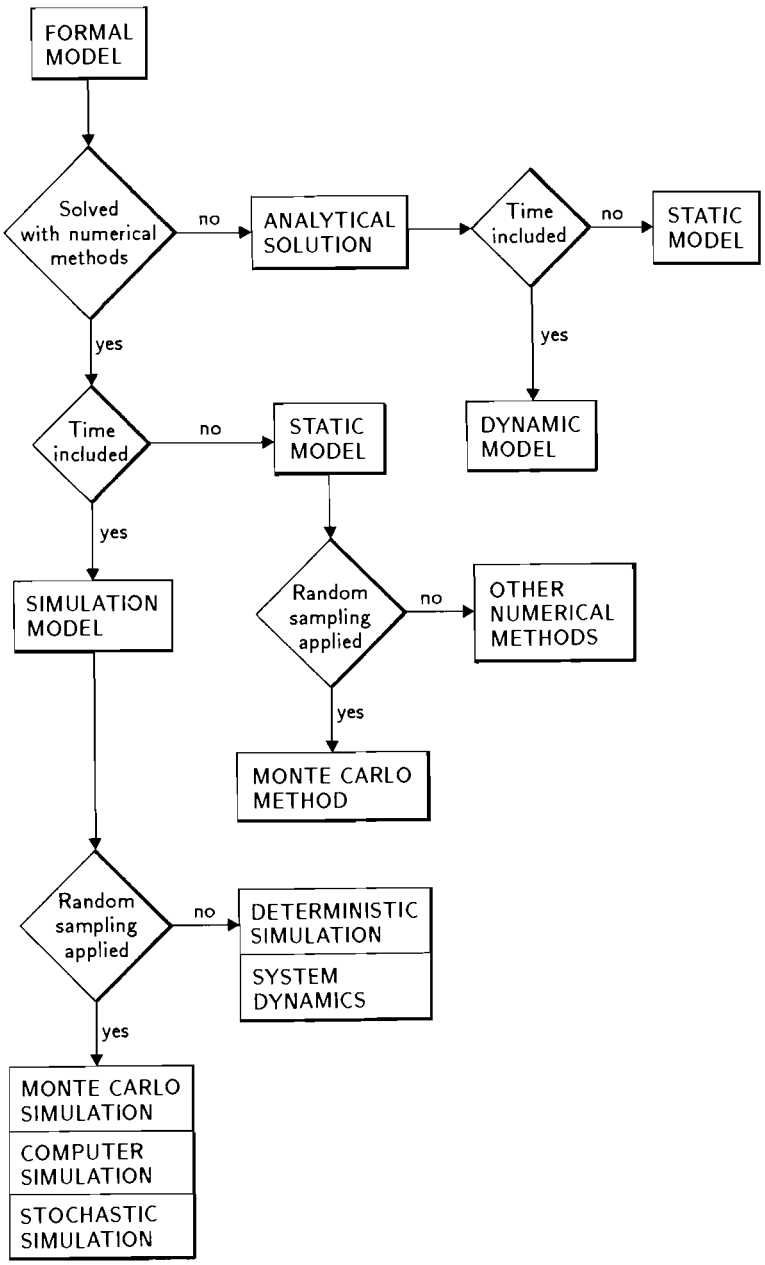

Figure 1.1 Flowchart describing types of models.

For example, a formalized model of the functioning of a telephone switchboard may consist of Boolean expressions that describe the state of switches; either 'on' or 'off'. However, a model that represents the functioning of the switchboard in terms of the frequency of calls that come in may not be adequately described by formal representations. Such a (simulation) model will require guidance by certain decision rules, that describe the consequence of specific events, i.e, what to do with calls when all lines are full. Such rules may be described by more flexible algorithms which for example may represent a model's output as result of randomly varying inputs (Mihram, 1974, p.8). The model will have to apply a random process to simulate the sequence by which calls enter the switchboard.

With the further development of systems analysis other characterizations of models have become relevant as well, such as:

1. the inclusion of time; a model with state values which change with time is called dynamic ,otherwise it is called static. According to Fishman (1973, p.12) a static model is only a snapshot of the state of a system at a moment in time. It should however be noted that a dynamic model that reaches a state of structural equilibrium may be considered as (quasi-) static, from that time on.
2. the solution method; a model is defined as deterministic when, according to Fishman (1973, p.12), a problem in a model can be solved analytically ,i.e. by means of mathematical relationships. Kleijnen (1974, p.5) notes that solutions for complex models can be achieved by numerical methods. These numerical solutions are obtained for each set of model parameter values (e.g., numerical integration). Fishman (1973) considers stochastic models to be the complement of deterministic models. In stochastic models at least a part of the systems behavior is random in nature.

The stochastic models of Fishman include, in contrast to Kleijnen, also simulation. Kleijnen (1974, p.5) considers two special numerical techniques, i.e., the Monte Carlo Method and Simulation. The term 'Monte Carlo' originated as a code word for the research on the nuclear bomb at Los Alamos in the U.S.A., around 1944. According to Hammersley and Handscomb (1964, p. 8) the credit for rediscovering the method goes

to Ulam, von Neumann and Fermi. One of the first scientific journal publications that explicitly describes the usage of the Monte Carlo technique for numerical integration, was written by Metropolis et al. (1953). In this paper "a general method suitable for fast computing machines, for investigating such properties as equations of state for substances consisting of interacting individual molecules is described". An exhaustive description of methods and techniques, including FORTRAN code for sampling from statistical distributions related to Monte Carlo analysis can be found in McGrath et al.(1973), McGrath and Irving (1973a, 1973b) and in Gardner et al. (1983). The Monte Carlo Method is defined "in a wide sense as any technique for the solution of a model using random numbers or pseudo-random numbers" (Kleijnen, 1974, p.6).

Simulation methods are, strictly speaking, distinct from Monte Carlo methods. Simulation implies experimentation, without necessarily using random numbers. There exist many kinds of simulation models like flight simulators, business and military games, and man-machine simulations. Kleijnen (1974, p.14) explicitly narrows simulation down to "experimenting with an (abstract) model over time, this experimenting involving the sampling of values of stochastic variables from their distributions". Kleijnen refers to this kind of simulation as stochastic simulation or, since random numbers are used, as Monte Carlo Simulation. Thus, the combination of simulation with random generators becomes a distinct modeling method (Figure 1.1).

Describing simulation in a wide sense as a technique of performing sampling experiments on the model of the system, Rubinstein (1981) derives the following four reasons for the application of simulation (see also Naylor et al., 1966, pp. 8-9):

1. it may be impossible or extremely expensive to obtain data from certain processes in the real world,
2. the observed system may be so complex that it can not be described in terms of a set of mathematical equations for which analytical solutions are available (i.e. economic systems)
3. even though a mathematical model can be formulated to describe some system of interest, it may not be possible to obtain a solution to the model by straightforward analytic techniques, and
4. it may be either impossible or very costly to perform

validating experiments on the mathematical models describing the system.

Rubinstein explicitly distinguishes between simulation, including stochastic simulation, and Monte Carlo methods. In contrast to simulation, Rubinstein states that (1) the Monte Carlo Method does not allow time to play as substantial a role as it does in stochastic simulation, (2) the observations in the Monte Carlo Method are independent, but serial correlation may be the rule in simulation models and (3) the response of a Monte Carlo Method may be expressed as a rather simple function whereas the response of a simulation model can be expressed only by the computer program itself.

Mihram (1972, p. 207) also distinguishes between the Monte Carlo Method and the simulation model on the basis of either exclusion or inclusion of time as an explicit required variable. Mihram states that "...even if one of the independent variables in the differential or integral equation would be interpreted as time, the Monte Carlo Method, when applied to its solution, seldom requires an explicit mechanism for advancing time or for keeping track of its passage". Mihram however also points out that "...distinctions that could possibly be made among the terms: sampling experiments, the Monte Carlo method, and Stochastic Simulation, will not likely be universally adopted" (Mihram, 1972, p. 208). Indeed, the wide definition of Monte Carlo Simulation, as used by Kleijnen, makes such distinction rather tedious.

Fishman (1973, p. 20) defines Monte Carlo Simulation as the random sampling part of the broader computer simulation which is the combination of "computer, operational rules, mathematical functions and probability distributions, given that a method of analytical solution is not readily available" (Fishman, 1973, p. 16). Since the definition of Fishman is merely adding a reason for applying stochastic techniques to the definition of Kleijnen, no distinction will be made further in this study between Computer and Monte Carlo Simulation.

The inclusion of probability concepts in simulation models need not necessarily lead to Monte Carlo Simulation. An economic model, for example, of which the coefficients have been estimated with regression techniques will provide a so-called deterministic simulation of the economic system. The prediction of the endogenous variables by exogenous and lagged endogenous variables does not include random

numbers any longer (Kleijnen, 1974, pp. 11-12). A more general example of deterministic simulation focuses on changes in system structure and policy to make the system better behaved. This kind of simulation has generally become known as system dynamics (Forrester, 1961; 1980).

Simulation and uncertainty

The advantages of introducing a probabilistic concept in modeling are, according to Mihram, threefold. Mihram (1972) first argues that symbolic models of the greatest complexity are becoming more and more common with the ongoing development of computing machines. Concentrating on digital computers "the capacity to keep track of large numbers of interacting components of a system meant that significantly improved modeling capabilities were available; no longer was the investigator constrained to represent the system's components interactions by formalized representations" (Mihram, 1972, p. 11). Second, the ability to represent random phenomena in the simulation model is important. The complexity of many models (e.g., a model representing the mixing of fluids) is such that a formalized model would have to describe attributes at a very disaggregated level (e.g., molecules), whilst the summarized probabilistic description of a set of molecules might suffice. "The internal relationships may be somewhat modeled by mathematical expressions, but a closer examination of the structure of the system reveals a much more profound and intricate internal behavior. This internal behavior usually comprises such a diversity of contributing effects that it is best described probabilistically" (Mihram, 1972, p.12). Thus, Mihram introduces probabilistic approaches as a relaxation of mechanistic modeling approaches especially when the confidence in the model results is not justifying the model detail.

The third reason, according to Mihram, for introducing a probabilistic concept in simulation is best described as insufficient knowledge of the investigator about the behavior of all system's components. "Deterministic models may be valid for the representation of system behavior on the gross scale .., but if such models do not readily compare with the observed behavior of the actual system, then a more detailed model may be in order. With the increase in detail comes

an increase in the likelihood of ignorance of the prevailing state of affairs and, concomitantly, of a stochastic model of the system" (Mihram, 1972, p. 14). This lack of knowledge is often referred to in terms of uncertainty. Uncertainty in the understanding of the investigator of the intricacies of systems is labeled by Mihram (1972, p.13) the "Uncertainty Principle of Modeling" which implies a broader notion that "refinement in modeling eventuates a requirement for stochasticity".

In systems analysis, the use of simulation models and probabilistic methods have increasingly been appreciated, not so much as a last resort for situations where no analytical approach was at hand, but rather as a means for reaching a better understanding of a system, its subsystems, their interactions and uncertainties.

A system as a set of subsystems; the aggregation problem

A complex system according to Mesarovic (1984) consists of relationships between components that are systems themselves.
Components of real world systems do not necessarily interact strongly with other components. Very often more or less independent subsystems may be distinguished; systems may be decomposable or nearly completely decomposable (Simon, 1961; 1978). The latter means that system relationships may be decomposed into individual blocks within which only strong interactions occur among the elements, while between such blocks only weak interactions may be distinguished. "This kind of hierarchical structure is often recursive with increasingly weak interactions as we go upward in the hierarchy" (Simon, 1978, p. 116). An economic system for example is built on the department-firm-industry-national and finally world market hierarchy. Similar hierarchies can be distinguished in the natural environment. A natural environmental system can be viewed as being built on the hierarchy of individual species that are contained in ecotypes, a set of which defines a natural system, and finally landscape and the biosphere, which consists of different natural systems.

The relevance of the concept of hierarchy in systems is that the (dynamic) behavior of a nearly-completely-decomposable-system may be analyzed without simultaneously examining each of the elements with

their attributes and their relationships. Depending on the problems that an investigator of a system wants to explore, a detailed system's description is not always required. If certain aggregated aspects of a system are of interest it may be that predictions can be obtained by using an appropriate aggregate model (Simon, 1978, p.114). The definition of 'appropriate aggregate' may, however, be influenced by the continuing controversy between holistic and reductionist modelers. The first group of modelers might favor a high aggregation level compared to the second, more mechanistically oriented, group.

In the process of modeling a system, the aggregation level of the model also depends on the data that has been sampled about the system and its environment. Decomposition of a system could consequently be guided according to criteria for grouping the data. The data could be clustered into homogeneous groups (see Rubin, 1967 for an overview of techniques) containing coherent[3] system elements and into heterogeneous groups such that intragroup homogeneity and intergroup heterogeneity are both high (Owsinski, 1983). Depending on the aim that is pursued, different kinds of groups could be defined. The (aggregated) model that is developed to simulate the behavior of the grouped system elements may thus become a function of the way the data are stratified. The composition of regions, for example, can - besides from a homogeneity principle - also be adapted to an existing administrative structure, to economic functions (Nijkamp et al.,1984) or to different ecotypes (Cale and Odell, 1979).

The aggregation concept can be extended towards the choice of the mathematical form of the model. In the modeling of monetary processes, for example, it has been shown that model-results are influenced by the modular structure and the relationships between modules (den Butter, 1986). It is therefore not surprising that it has been demonstrated that a simple linear relationship instead of a non-linear model may represent the same set of system dynamics (see for example O'Neill and Gardner, 1979; Gardner et al., 1982; Cosby et al.,1985b, Hornberger and Cosby, 1985b, Hettelingh and Gardner, 1988). The aggregation to simple mathematical relationships may involve the establishment of a relation between the outputs of a (computer-) model and its inputs, treating the

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[3]A group of elements is called 'coherent' in the view of Owsinski (1983), depending on the definition (i.e., the measure) used to describe the distance between the elements.

model as a black box, which is referred to as the response surface methodology (Downing et al., 1985; Iman and Helton, 1988). When the relation between inputs and outputs of a simulation model are established by means of regression analysis the term metamodeling, introduced by Kleijnen (1987), may be employed.

In this study, metamodeling plays an important role in the design of experiments to investigate the merits of zoning for environmental policy making.

Systems analysis and policy making

Systems Analysis is useful for policy making if the results of such analysis, as they pertain to a particular policy, are clear. However, clarity is dependent on the harmony between the policy problem, the isolation of the system involved, the choice of the proper aggregation level, and the confidence that may be assigned to the analysis results. Therefore, uncertainty analysis is of vital importance to judge the credibility of model results. The extent to which results are to be trusted is nevertheless contaminated by many kinds of errors, e.g., due to uncertainty of system perception, by insufficient and erroneous data, and in model structure and computer programs. These errors may propagate through the model (see Clifford, 1973) that yield unexpected results of dubious credibility for, among others, policy makers.

The present study cannot provide a complete overview of the advantages and pitfalls of systems analysis to policy making. The scope has been restricted to our natural environment. The reasons are the current policy relevance, the scientifically challenging complex features and the uncertainty involved in modeling parts of the consequences of human activities on the environmental system.

1.3 ENVIRONMENTAL MODELS FOR POLICY APPLICATIONS

The attention addressed to our natural environment has increased over the past years due to the exhaustion of various natural resources, the damage to forests and lakes by acid rain, the legal and illegal

dumping of chemical waste, the controversy about nuclear power and nuclear waste disposal, the nuclear power plant failures at Harrisburg (U.S.A) and at Chernobyl (USSR), the desertification in Africa and deforestation in tropical forests, and, finally, the recently projected changes in global climate. The immediate cause of these environmental and resource problems is the uncompensated social costs associated with the use of ecological systems for socioeconomic production and consumption activities.

The many interacting systems that lead to the decrease of the vitality of environmental systems can be treated in a mono-disciplinary way provided that the temporal scale is short or the spatial scale is small, and otherwise should be approached in a multi-disciplinary way (see also Braat and van Lierop, 1987a, p. 8). Multi-disciplinary approaches to environmental systems have led to integrated environmental modeling (see Brouwer, 1987 for an overview). The validation of environmental damage (Opschoor, 1974) and the modeling of integrated economic and ecological system processes has in the past years received great attention (see also Nijkamp and Opschoor, 1977; Lakshmanan and Ratick, 1980; Arntzen et al., 1981; Brouwer et al., 1983; Hafkamp, 1984; Brouwer, 1987). An integrated approach to environmental model building called Adaptive Environmental Assessment (AEA) has been introduced by Holling (1978) and Walters (1986) which aims at bringing together different disciplines for brief periods of intensive workshop interactions. Such modeling workshops consist of a team of mathematical modelers, research scientists from various disciplinary backgrounds, resource managers with experience in monitoring and regulation, and finally policy analysts or decision makers with a broad responsibility for defining management objectives and options.

In recent years there has been a tendency to provide the policy maker with information management systems (see also Nijkamp and Rietveld, 1984), integrating data, models and expert opinions about the natural environmental system and its environment. An example of an approach towards such a model (Braat et al., 1987) on a European scale deals with natural ecosystems, fisheries, forestry, agriculture, air- and water pollution, and economic and energetic processes. The validation of environmental policy through user friendly computer programs, where results are displayed graphically and user inputs are

driven by simple menus, is another recent development. The Regional Acidification INformation and Simulation (RAINS) model, which is developed at the International Institute for Applied Systems Analysis (IIASA), is such a tool which integrates modules that compute European sulfur and nitrogen emissions with modules evaluating deposition, forest soil and surface water acidification (Hettelingh and Hordijk, 1986; Alcamo et al., 1987; Hettelingh and Hordijk, 1987). Because the present study draws on parts of RAINS, a detailed review of this deterministic simulation model will be given in this study.

The usage of this kind of models, and concepts by policy makers, is holistic in nature; the models are not used to simultaneously evaluate all socioeconomic and environmental mechanistic consequences, but are used to evaluate pre-targeted criteria like 'Lowest Admissible Emission Rates' (LAES) or 'Prevention of Significant Deterioration' (PSD)-levels (see also Wetstone and Rosencranz, 1983 for policy legislation in the U.S and Europe regarding acidification). In March 1984, Ministries of nine countries agreed on reducing sulfur-dioxide emissions by 30% in 1993 based on national emissions in 1980 (see also Hordijk, 1986). This kind of policy thresholds is generally called target loads which are used in targeted policy strategies aimed at goals or constraints implied by so called indicators (see Batterman et al., 1988, p.543). Indicators may represent economic factors, like 'Reasonably Available Control Technology' (RACT) and investment costs, or be based on environmental impacts, i.e dose-response functions (see also Opschoor, 1974). The concept of impact indicators (Munn, 1975) as an aggregate measure of the condition of an environmental subsystem has become increasingly operational to policy making (see also Vos et al., 1986). It is clear that target loads that are based on technical or economic considerations, may still cause unacceptable damage to the environment. Therefore, the highest load that will not cause chemical changes leading to long-term harmful effects on the most sensitive ecological systems (i.e. critical loads) are becoming increasingly important for evaluating acid deposition effects (Nilsson, 1986).

The concept of indicators in general and critical loads in particular is still controversial. It categorizes the quality or state of parts of our environmental system, without the need to fully

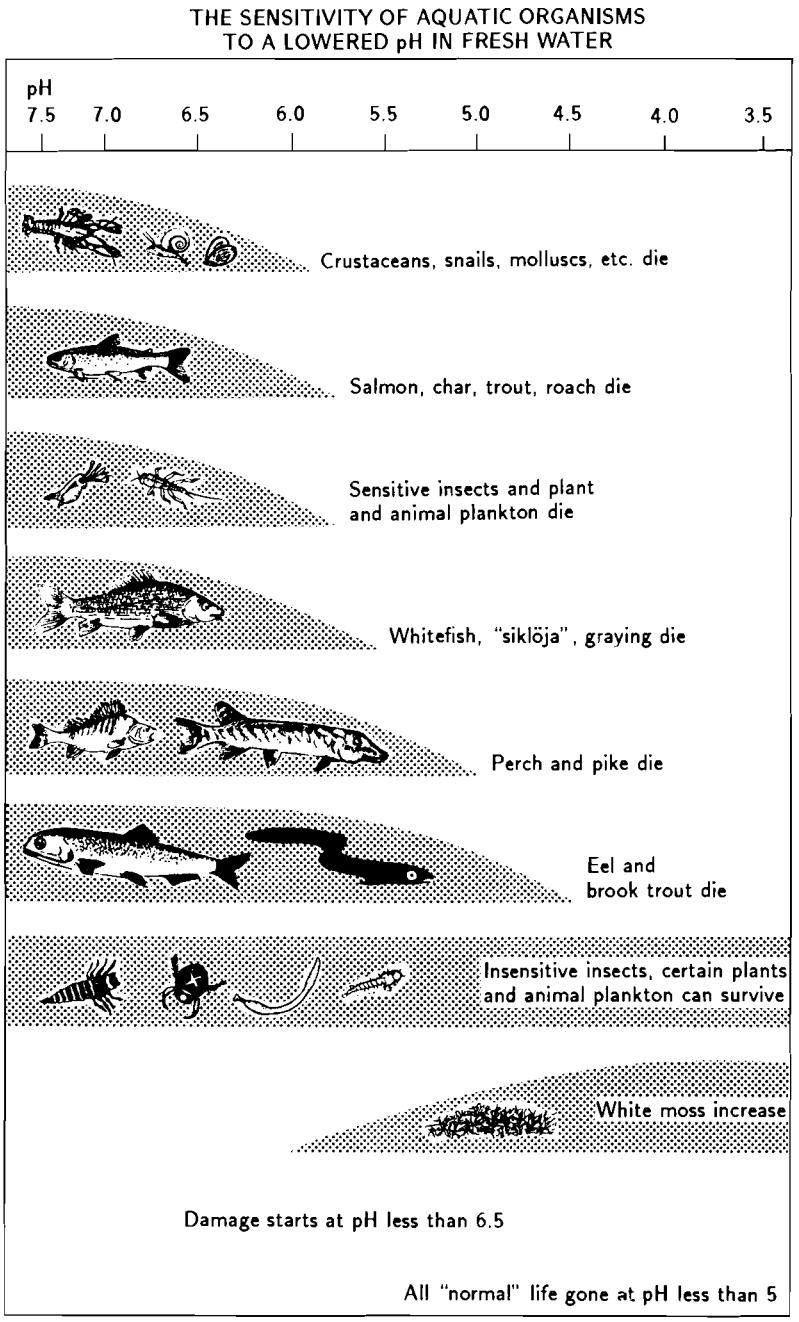

Figure 1.2 The sensitivity of aquatic organisms to a lowered pH in fresh waters (Source: Ministry of Agriculture of Sweden, 1982).

understand causes and effects of all systems that may be directly and indirectly involved. It is a holistic approach to systems in which the indicator is assumed to represent the state of the system as a result of the relations to other system elements. An example of this kind of categorization of the effect of watershed acidification is shown in Figure 1.2. In Figure 1.2 the extent of acidification of a watershed is described in this Figure by pH levels (see chapter 3, equation 3.4). At different pH levels, the extent of the damage to the flora and fauna in watersheds is shown to increase.

Without the understanding of systems, however, managers and policy makers would probably not be able to face the increasing demands to produce quantitative predictions of environmental responses to disturbances, such as air pollution.

1.4 SCOPE AND OUTLINE OF THE STUDY

This study will identify key factors determining watershed responses to acid (sulfur) deposition, in different regions. The study aims at

1. providing a method of defining a region such that environmental policy directed at alleviating watershed acidification, becomes more suited for large scale management, of surface water quality
2. depicting regional characteristics that allow for the usage of a less detailed, thus more aggregated model, and
3. providing an operational concept of critical loads for policy insight into watershed quality as a function of the spatial zoning of watersheds.

To keep the study manageable, an existing model has been used to illustrate the above goals. The model that has been used predicts acidification of surface water, i.e. lakes and watersheds. The model is part of the Regional Acidification, INformation and Simulation (RAINS) model, developed at the International Institute for Applied Systems Analysis (IIASA), and is therefore called the RAINS Lake Model.

ad 1. Regionalization or zoning, as pointed out earlier, often

entails some kind of aggregation. This aggregation may often not be suited for the problem that is investigated; a region contains many different kinds of lakes with respect to the response to acid deposition. The application to a region of a mechanistic[4] description of the acidification of one lake for the purpose of policy making, e.g., evaluating the state of acidification as a function of critical loads, may lead to biased conclusions with respect to some of the lakes in that region. Rather than investigating the average state of lake acidification in a region, policy should be focussed on the state of lake classes within that region.

A new method of flexible zoning is introduced that allows for the policy assessment of environmental systems in regions.
The method is aimed at reducing the interdependency between environmental model predictions and the choice of the zoning system.

ad 2. The treatment of many lakes that are zoned into lake classes, e.g. sensitive and insensitive lakes, also has consequences for the aggregation level of the model used to describe regional lake acidification. The necessity to mechanistically approach sets of lake acidification systems, depends on the uncertainty of model predictions as a function of zonal system components, viz., soil characteristics and deposition patterns. The correspondence between metamodel response and the RAINS Lake Model predictions is used to investigate the homogeneity of lakes of which the acidity is simulated within a zone.

ad 3. The policy exercise of matching regional target and critical loads deterministically may, as mentioned earlier, nevertheless lead to damage to parts of the environmental system e.g., particular lake classes. A more probabilistic approach towards the estimation of sets of lakes under risk will be proposed in this study as an alternative.

The questions to be addressed in the study are, as mentioned before, illustrated using the RAINS Lake Model. The complexity of the RAINS Lake Model, the scarcity of input data and the importance that will be attributed to uncertainty are reasons for Monte Carlo Simulation to be used throughout this study.

The study contains 7 chapters. The first three chapters review the

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[4] A mechanistic model is the result of a reductionist approach to systems (see section 1.1).

field of environmental modeling in general, the application of uncertainty analysis techniques, and watershed quality modeling in particular. The following three chapters consist of the design and results of the investigation to quantify the uncertainty aspects of the regional RAINS Lake Module. The last chapter summarizes the study.

The first chapter has introduced the nature of the study and relevant terminology, although some terms (e.g. uncertainty, Monte Carlo) are treated in more detail in the following chapters. Chapter 2 gives a review of techniques that have recently been developed in the field of environmental modeling, while also approaches related to environmental model structures are considered. Then, attention is given to the uncertainty involved in modeling an environmental system. Next, modeling requirements with respect to input data and the need for model calibration as a function of available data, in relation to the Monte Carlo method is treated. These modeling requirements are also related to the potential that the model output provides to policy makers. This chapter concludes the part of the study where environmental modeling is treated from a systems theoretical point of view.

Chapter 3 explains the processes in the soil and lake, due to acid deposition, that lead to lake acidification . An overview of state-of-the-art lake acidification modeling in general and the RAINS Lake Module in particular is presented next.

Chapter 4 presents the research design and the results of applying the RAINS Lake Module on predefined regions in Scandinavia using standard emission scenarios from RAINS through 2040. It will be shown that the calibration methods needed to obtain behavioral significance of the model applied to a particular zoning system, i.e. predefined regions, may lead to model predictions that are less appropriate for targeted policies.

In chapter 5 the method of flexible zoning is developed and illustrated to distinguish between lake classes within a region. The use of mechanistic vs. metamodels is treated in the light of model uncertainties and lake classes. It is shown that the concept of critical loads can be operationalized on the basis of lake classes within a region under consideration.

Chapter 6 describes the results of applying the method of flexible zoning on Dutch watershed data. The Dutch area is overlaid by four acid deposition level classes within which a total of eight watershed zones

are distinguished. Model predictions are obtained for each of these watershed zones in combination with the acid deposition level classes just mentioned and in combination with critical loadings.

Finally, Chapter 7, evaluates the study and its potential: the early inclusion of uncertainty in the application of a model to regional systems improves the understanding of the compatibility between model predictions, the choice of regional boundaries and available data.

2. ISSUES IN ENVIRONMENTAL MODELING

2.1 INTRODUCTION

Environmental modeling is a subset of modeling in general. Like in any other effort to represent a system by means of a formal model, environmental modeling is concerned with the analysis and identification of relevant systems processes. Our natural environment is, however, not isolated from the systems in which daily life is taking a course, so that not one single system needs to be understood, but many interacting ones. Models, including integrated models are, according to Mihram (1972, p.213) built in five stages:

1. Systems analysis, which isolates a sufficient set of state variables to represent the system and the interactions among the elements of the system.
2. Model synthesis, which includes the delineation of the necessary state variables, entities, attributes, relationships and input data. It also requires the necessary computer routines to be written.
3. Model verification, which consists of comparisons of the model's behavior and the behavior that was intended in the programming structure. This stage consists of the correction of the model syntax as a result of experiments (see also Kleijnen, 1974, p. 75). For example, checking if the sign (- or +) of the model response, under varying inputs, corresponds to what is expected, is a typical verification experiment.
4. Model validation, which consists of comparisons of the responses of the model, that was verified in stage 3, to observations taken on the modeled system.
5. Model analysis, which consists of the experiments with the verified (stage 3) and the validated (stage 4) model to address those questions which initially prompted the construction of the model.

The multi-disciplinary nature and the large temporal and spatial

scales of environmental systems make the distinction of these stages dependent on the hierarchical arrangement of a system (see chapter 1). The definition, for example, of boundaries of an environmental system is heavily dependent on one's view of the way in which elements of the nature interact, i.e. the causal relationships within the natural system. Although synthesis and verification of the model, that is developed within the frame of these boundaries, may be successful, validation may remain a problem due to causalities that were excluded as a consequence of the boundaries chosen. An additional problem is the independence between data sampling setups, also referred to as monitoring programs, and the input requirements of models that may even have been non-existent at the time when the data were collected. Finally the time and spatial scale at which processes of environmental systems take place are often too broad to allow for validation by means of data collected at a few geographic locations at a particular point in time (see Brouwer, 1987). In general, there is a considerable discrepancy between the data needed to validate environmental models and the available data. This also has consequences for the stage of model analysis. Therefore, in environmental modeling the need for assuring that model predictions resemble observed patterns, is becoming an important factor in the validation stage. This, for example, is done by means of adaptation of parameter values in an iterative sequence of model runs. This adaptation exercise is often referred to as calibration (see also Kleijnen, 1987, p.186). More attention is also needed for calibration in relation to the treatment of uncertainties in model parameters and in model relations, and of errors in field data measurements (see also Beck, 1983).

This chapter will summarize some recent concepts of environmental model structures and present some examples of recent environmental models. Much attention will be given to the RAINS model because of its relevance for this study. The last three sections of this chapter will be aimed at familiarizing the reader with the analysis of model uncertainties and the related topics of calibration and stochasticity. Since stochasticity of model behavior and model response is closely related to Monte carlo Simulation, a rather detailed statistical expose of random sampling techniques is included in the last section of this chapter.

2.2 MODEL STRUCTURE: INTEGRATED ENVIRONMENTAL MODELING

In chapter 1 an overview was given of different causes that have recently led to increased focus on our natural environment. The global changes of our biosphere, e.g. climatic change, has urged the need for a simultaneous treatment of causes and effects; the human activities with respect to industrial and resource development all are interacting causes of the depletion of the environmental quality. The effects vary from rather local household waste disposal problems to global changes, e.g. damage to the ozone layer by chlorofluorocarbons set free by dumped refrigerators. The simultaneous treatment of linked processes in different systems has therefore increasingly become a tool to analyze environmental changes.

Following Hafkamp (1984, p. 25) an Integrated Environmental Model (IEM) is defined as a type of model in which a great diversity of real-world phenomena, including those related to the natural environment, and their inter-dependencies are represented simultaneously. According to Hafkamp (1984, p. 25), an IEM may encompass the modeling of:

1. environmental impacts, inter alia due to air pollution (e.g. acid rain as result of sulfur and nitrogen compounds), solid and toxic waste;
2. ecological systems: terrestrial and marine ecosystems, predator-prey systems etc.;
3. resources management: exploitation of energy sources (i.e. coal, oil, gas), forests;
4. economic systems: macro, meso, micro scale systems;
5. social and political systems: social layers, activity pattern (e.g. consumer organizations), unemployment rates;
6. transportation systems: (rail)road networks, airport and harbor facilities;
7. demographic developments: age structure, educational profiles.

In general IEMs are characterized by a multi-disciplinary approach to relevant phenomena of the natural environment (Brouwer, 1987, p.11).

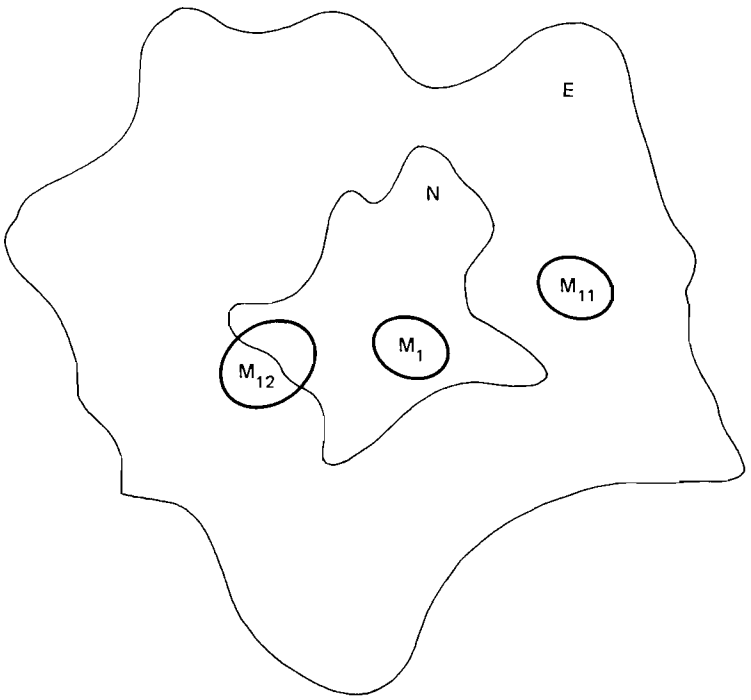

Figure 2.1 IEM modules representing parts of the real world system. N represents the set of natural environmental systems. E represents the set of other systems. M1, M11 and M12 are modules of the, as yet not represented, model M which controls the relationships between the modules.

Linked components of different subsystems, e.g. the seven examples listed above, can be modeled by linking various sub-models, called modules. A module will be interpreted as a set of intra-related, often mono-disciplinary oriented, variables (Brouwer, 1987, p.11), that represent phenomena of a subsystem and that are inter-related to variables of another module. In an IEM, one of the modules will be representing a part of the natural environment. A pictorial representation, of an IEM is given in Figure 2.1, which displays the relation between a model and its sub-models; computations of M1 can only be performed with inputs provided by M11 and M12. Important for the system's understanding and the model design is the way by which the modules are linked. The linking structure of such, often mono-disciplinary, modules may be:

1. horizontal: all disciplines involved are regarded as equal constituents for building a multi-disciplinary model (Nijkamp, 1987, p.30),
2. vertical: one discipline is regarded as superior to the others, so that the relationship from the dominant discipline to the remaining ones receive special attention (Nijkamp, 1987, p.30),
3. satellite: a mixture of a horizontal and vertical structure (Figure 2.2), organized around a core module (Brouwer, 1987, pp. 79-80)
4. multi-layer: more layers represent the same set of subsystems, but the disciplines by which the subsystems are modeled vary amongst the layers (Hafkamp, 1984, p. 69). Thus, intra-layer relations between similar modules of each layer, vary from layer to layer. Inter-layer relationships ensure multi-disciplinary relations between the modules that represent the subsystems. An example of a triple layer approach (Hafkamp, 1984, p.77) is presented in Figure 2.3.

In recent years, many IEMs have been constructed. An overview of IEMs can be found in Hafkamp (1984), Brouwer (1987) and in Braat and van Lierop (1987b), and hence not reviewed in detail here. To avoid too vague an understanding of the concept of integrated modeling, three recent examples are presented below, viz., the Modeling and Information System for Environmental Policy (see also Hettelingh et al., 1985),

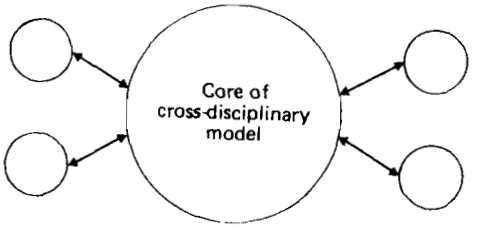

Figure 2.2 A satellite structure for a cross disciplinary model (source: Nijkamp, 1987, p.31)

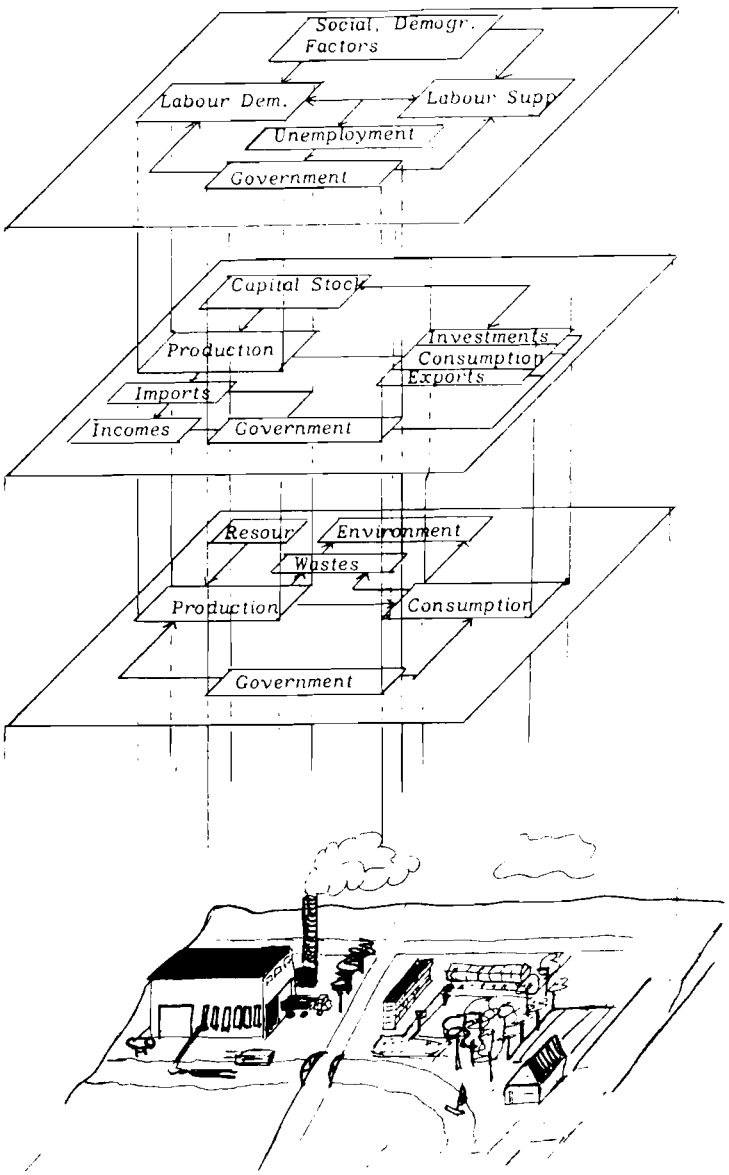

Figure 2.3 A simple version of the Triple Layer Model obtained by multi-layer projection in three layers (source: Hafkamp, 1984, p. 77).

the Integrated Regional Environmental Model for Physical Planning (Arntzen et al., 1981; Brouwer et al., 1983), both operational on respectively a national and regional scale in the Netherlands, and the Regional Acidification, INformation and Simulation model, RAINS (Hettelingh and Hordijk, 1986; Alcamo et al., 1987), which describes and predicts European acidification. The similarity between the models is the chained treatment of the cause and effects of environmental stress. Dissimilarities occur with respect to the way in which the models are driven (e.g. where does the model start), and to its transparency for policy purposes (e.g. user-friendliness; prediction confidence).

The Modeling and Information System for Environmental Policy (RIM)

The RIM is a Dutch model that integrates economic trends and constraints, processes of technological change and innovation, pollution abatement policy, and population dynamics whilst it provides input to long term dynamics of ecological processes. The general structure is presented in Figure 2.4.

The RIM model is based on use of extended input-output tables. The major addition to the traditional analysis of pollution via input-output tables (e.g. Leontief, 1970), is the detailed description of economic and energy flows into the tables. By relating emission factors to these flows of energy products and other goods, the extended input-output tables enable the construction of a detailed mass balance table for every pollutant. The tables are constructed on the basis of official Dutch economic projections, enabling the estimation of input output tables for all the target years encompassed by the economic projections.

If the structure of the model were to be defined in terms of the definitions presented above, the vertical structure applies because of the superiority of the projections of the economic system as compared to other systems involved in driving the model. When not so much the model driver, but rather the organization of the input-output relationships serve as basis to define the model structure, then the

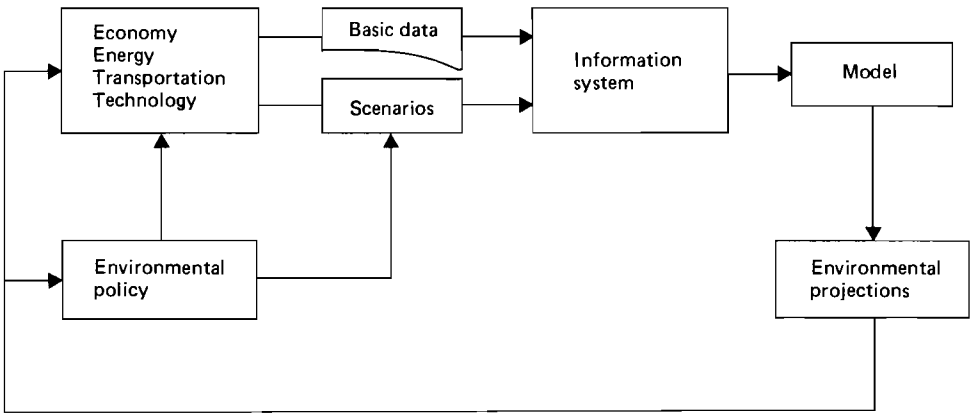

Figure 2.4 The general structure of RIM

multi layer structure may apply as well; the great many tables, i.e. economic Input-Output tables, emission Input-Output tables of different pollutants may be layered on top of one another.

The Integrated Regional Environmental Model for Physical Planning (GMM)

The purpose of the GMM was to simultaneously analyze socio-economic change, spatial planning and effects on the (regional) natural environment, triggered by a policy intention to transform a small water area into land. A case study (Arntzen et al., 1981; Brouwer et al., 1983) was carried out for a small region in the southern part of the Netherlands.

Five modules were designed (Figure 2.5):

1. The demographic module describes the composition and development of the population.
2. The economic module quantifies production, land use and air pollution. The module is driven by a consistent, interrelated plan of the future in which employment is the crucial variable. The employment variable is a function of the population composition, derived in the demographic module.
3. The facilities module quantifies recreation, water use, waste, housing and air and water pollution caused by households. This module is linked to the demographic module through total population, to the economic module through employment and to the ecological module through the recreation.
4. The ecological module identifies the main homogeneous natural environmental regions, called geotones. Two such geotones were discerned, one of which was treated in a purely qualitative way because of lacking data. The development of the nature is a function of stress incurred by recreation, households and economic activities.
5. The intermediate module contains the dispersion module of sulfur-dioxide in the air using a Gaussian plume model, and takes care of the bookkeeping to balance land supply with the demand.

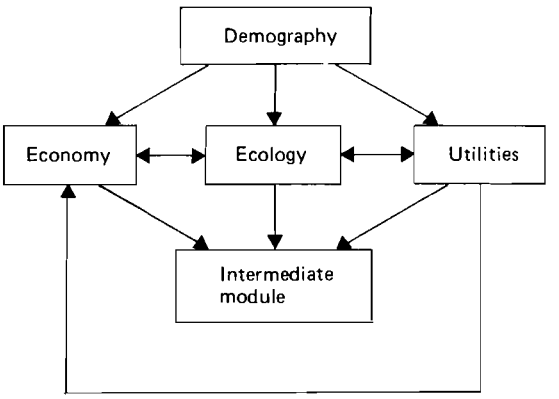

Figure 2.5 The structure of the modules of the GMM.

The GMM is driven by the demographic developments that feed the economic scenarios.

Because of the importance of the demographic module, the vertical structure seems the most appropriate definition, as far as the model driver is concerned. The central position of the economic module would also allow for the satellite structure to be used to describe the GMM setup.

A definition of the structure of a model, as can be seen from the examples given above regarding the RIM and the GMM, depends on the way in which the model and its functional relationship is viewed. The main advantage, as pointed out earlier, of defining a priori model-structures is the resulting frame of thoughts, provided to an investigator of a system. However in terms of model applicability it seems that a priori concepts of model structure are less important than the stage of validation, verification and model analysis (section 2.1), which directly affect model results.

Finally it can be remarked that both models are deterministic, have not yet been subjected to uncertainty analysis, whilst their utilization is dependent on qualified investigators in the sense that neither the GMM nor the RIM are provided with user-friendly interfaces.

The RAINS model for the prediction of European acidification

The RAINS model (Figure 2.6) is in many respects on top of the current mainstream of the development of IEM's; it uses deterministic as well as stochastic techniques including calibration, it is highly interactive and within reach of many types of users, and many of its modules have been made subject to sensitivity and uncertainty analysis (Hettelingh and Hordijk, 1986; Alcamo et al., 1987; Hettelingh and Hordijk, 1987). In the following the description of RAINS will be restricted to its main features.

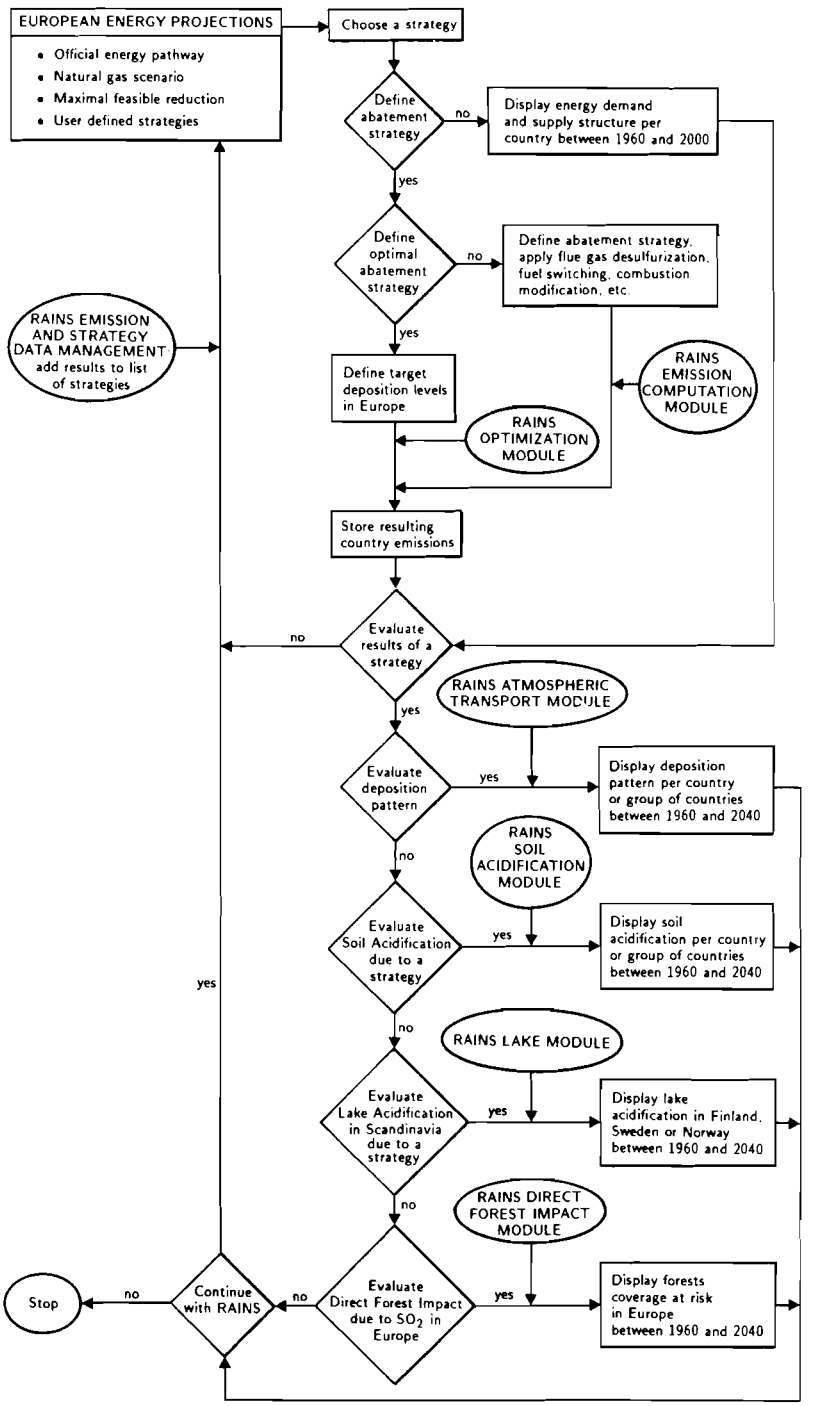

Figure 2.6 A simplified flowchart representation of the Regional Acidification, INformation and Simulation (RAINS) model.

The subsystems that have been modeled and linked in RAINS are (1) the energetic[5] (2) the atmospheric transport (3) the soil acidification (4) the direct forest impacts of SO_2 concentrations and (5) the lake acidification system. The integrated treatment of causes and effects by RAINS consists of a detailed description and prediction of fuel combustion in the European countries (causes) and the effects on European forest soils and Scandinavian lakes. The combustion of many fuel types leads to the emission of sulfur and nitrogen oxides of which the dispersion over Europe is simulated in RAINS by means of a source-receptor matrix. Such a matrix describes the deposition anywhere in Europe due to emissions in European countries. The matrix has been derived from an atmospheric transport model (Eliassen and Saltbones, 1983). Such a model takes the non linear relationships into account between stack heights, atmospheric mixing layers, wind fields and many other atmospheric processes.

The effects submodules of RAINS are integrated models as well, e.g. the soil and lake acidification modules describe different systems like water flows over the surface of, and within different layers of the soil. In each soil-layer chemical reactions are simulated between deposited acid and soil-minerals. RAINS thus integrates large scale, macro processes (e.g country emissions, depositions) with small scale, micro processes (e.g. depletion of soil minerals by acid deposition). The problem in this kind of macro and micro process integration is how to make large scale predictions that are based on micro processes. As mentioned earlier the RAINS Lake Acidification Module is used to investigate this problem (chapter 3 and following).

Besides the various scales at which the RAINS submodels operate, also the scales at which RAINS results are displayed vary. The spatial scale of the RAINS model is Europe, varying from approximately 4250 km^2 large grid squares in the deposition and soil acidification module, 14 so-called lake regions in the lake acidification module, to 27 countries in the energy-emission computation module. Its temporal scale covers the period from 1960 to 2040.

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[5]The economic system has not been modeled explicitly. The modeling of European energy demand and supply structure is based on official governmental projections.

The RAINS computation modes

The RAINS model (Hettelingh and Hordijk, 1986; Alcamo et al., 1987) basically performs its computations in three steps (see Figure 2.6 for a more detailed partitioning):

1. Energy use patterns, classified by 8 fuel types and 6 energy sectors, are the bases to compute national emissions of sulfur and nitrogen oxides. Users may define different emission patterns by applying pollution abatement techniques, of which also the costs are evaluated. The module provides graphical and tabular displays of user defined pollution abatement strategies.
2. Deposition patterns of sulfur and nitrogen are computed by multiplying country emissions by a source receptor matrix (Eliassen and Saltbones, 1983).
3. Environmental effects of sulfur are computed as a function of the deposition in European forest soils and Northern European lake regions. Also direct forest impacts of sulfur air concentrations are predicted.

RAINS consists of computational modules as well as display modules. The soil and lake acidification, for example, can only be displayed after translating sulfur deposition ($g/m^2/yr$) in a measure for acidity, the pH (see chapter 3).
RAINS is driven in two ways i.e. in a scenario analysis mode and in an optimization mode:

1. scenario analysis consists of the user definition of sulfur and nitrogen abatement strategies as function of scenarios that embody the official government predictions of the energy use until the year 2000 for all countries throughout Europe, and
2. optimization consists of the definition of minimum cost abatement strategies subject to a target deposition in a geographic location anywhere in Europe (see also Batterman et al., 1988).

RAINS used for policy-making

RAINS is increasingly becoming an important tool for evaluating European policy aimed at abating sulfur and nitrogen pollutants. Such policies can not be restricted to a local or national scale because of the fact that country emissions lead to depositions in other countries. Abatement policies, therefore, lead to transboundary alleviation of environmental damage. In july 1985 a protocol was added to the Convention on Long-Range Transboundary Air Pollution (Economic Commission for Europe (ECE), 1985) , signed by 32 countries, the European Economic Community (EEC), Canada and the USA, stating that sulfur emissions or transboundary fluxes will be reduced by at least 30% (ECE, 1987) as soon as possible and at the latest by 1993 (using 1980 levels as the basis for calculation of reductions). Recently a similar protocol on nitrogen oxide emission reductions was signed. This protocol (ECE, 1988) states that by 1994 emissions of nitrogen oxides should not exceed the level of a year, to be chosen by each country, before and including 1987. In addition the average emission between 1987 and 1994 should not exceed the 1987 emission level.

In february 1989 the ECE "Task Force on Integrated Assessment Modelling" (TFIAS) who is coordinating the development of integrated assessment modeling in Europe, has recognized RAINS as the only regional acidification model that is capable of developing and assessing European strategies. The "Working Group on Abatement Strategies" , to which the TFIAS reports, has officially recognized RAINS as the primary tool to be used by the ECE for developing the new sulfur and nitrogen oxides that will succeed the protocols mentioned above.

Some examples of RAINS results

In the Figures 2.7 through 2.11 examples are presented that illustrate the way by which RAINS compares two alternative policy strategies. Figure 2.7, displays the deposition pattern of Europe in 1995 when no abatement is applied. The country emissions causing this pattern result from the governmental projections of energy use until 2000, which is denoted in RAINS as the Official Energy Pathways. The

result of a 30% reduction on 1980 emissions has been used as input into RAINS as a strategy called Current Reduction Plans of which the deposition in 1995 is displayed in Figure 2.8. It is clear from Figure 2.8 that the area with depositions of more than 10 $g/m^2/yr$ in central Europe is reduced, compared to Figure 2.7. The same area has become subject to a deposition in the range of 8.0 to 10 $g/m^2/yr$. The effects for soil acidification of the Current Reduction Plan strategy is shown in Figure 2.9. The area with a pH smaller than 4.3 is becoming smaller with this strategy as compared to the area with a similar soil pH in the Official Energy Pathways over the period from 1985 to 2040.

Taking into account the delayed response of the nature to changes in external stress, the state of lake acidification is displayed in Figures 2.10 and 2.11 for 2040. In Figure 2.10 the effect of the Current Reduction Plan strategy is shown, while the consequences of applying all available technologies in Europe to reduce emissions is displayed in Figure 2.11.

This Maximal Feasible Reduction strategy takes into account what the country potential is to invest in pollution abatement equipment. The difference between the effect of the two scenarios is a reduction of 1.8%, leading to about 8.1%, of lakes with a ph lower than 5.3, of 2.2%, leading to about 16.1%, of lakes with a ph in the range between 5.3 and 6.3 and an increase with 4%, to about 75%, of the lakes with a pH higher than 6.3.

The RAINS Lake Model prediction of lake acidification in northern Europe is obtained by using deposition levels of years beyond 1980 as input into the model. Additional parameter values needed to run the model are the same as those used in the model when it was validated to 1980 pH measurements. In other words the model was calibrated (see section 2.4) in the period from 1960 to 1980 to provide predictions in the period 1980 to 2040. The calibration procedure was repeated for 14 predefined regions in Scandinavia. The confidence to be attributed to the model predictions thus is dependent on the accuracy of the calibration and may, as pointed out before, be a function of the zoning of Scandinavia to 14 subareas. The zoning has led to an aggregation of lakes each of which may differ significantly from one another, i.e. some may be big and insensitive to acid deposition, others small and sensitive. It may very well be that a particular abatement strategy leads to only the sensitive lakes to become acidified.

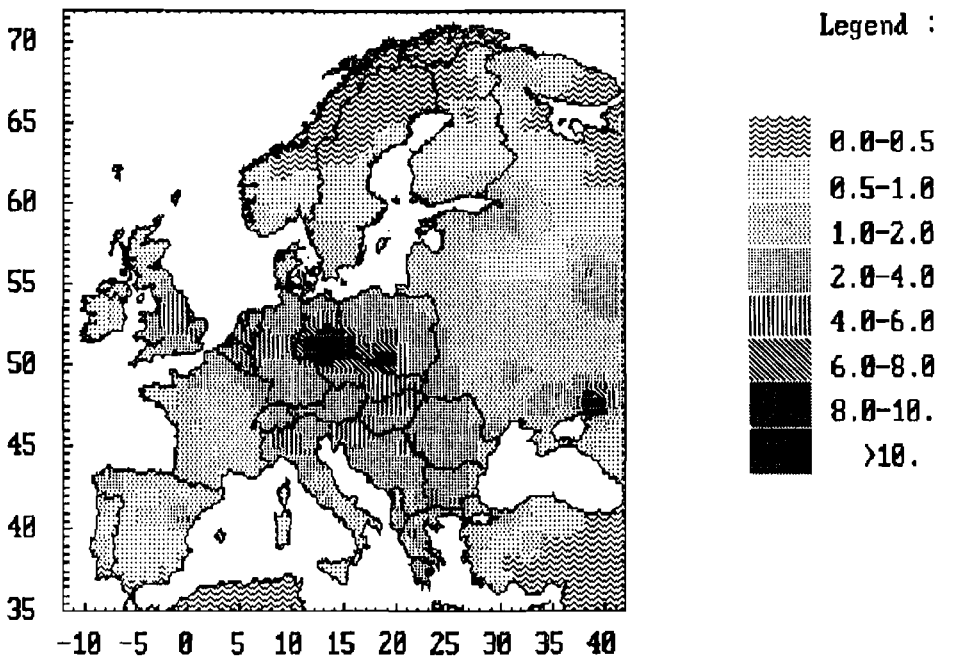

Figure 2.7 Deposition pattern of Sulfur ($g/m^2/yr$) in 1995 resulting from country emissions as projected by the governments, referred to by RAINS as the 'Official Energy Pathways'

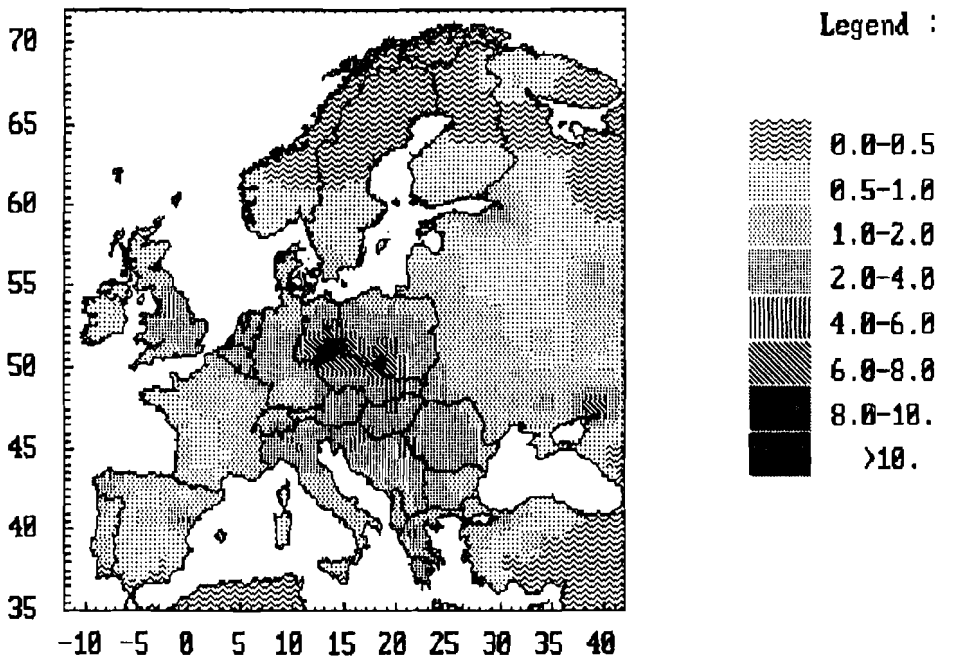

Figure 2.8 Deposition of Sulfur ($g/m^2/yr$) in 1995 due to country commitments to reduce at least 30% of the 1980 emission levels, referred to in RAINS as 'Current Reduction Plans'.

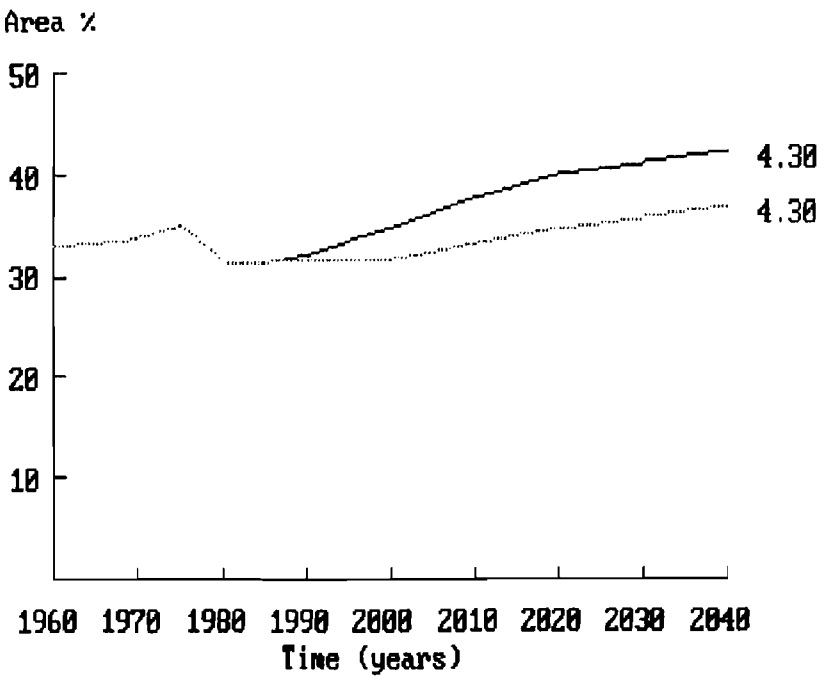

Figure 2.9 Time evolution of forest soil areas in Europe with a pH below 4.3 due to the 'Official Energy Pathways' (solid line) as compared to the 'Current Reduction Plan' (dotted line) scenarios.

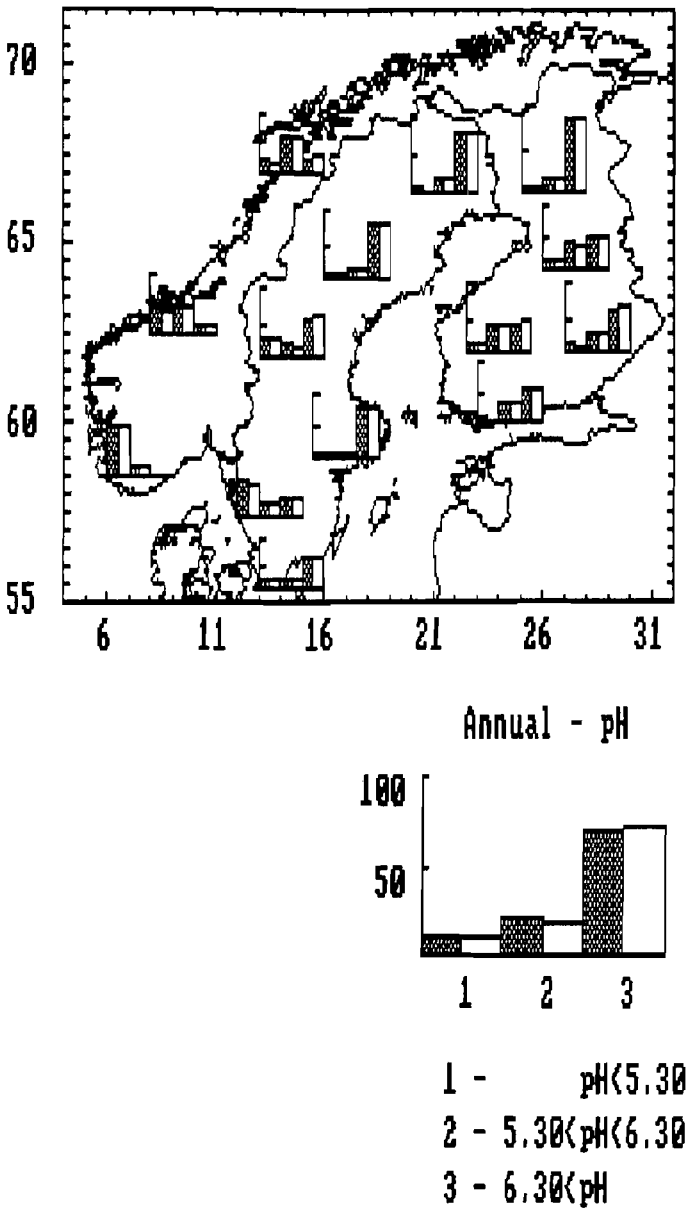

Figure 2.10 State of lake acidification in 14 predefined lake regions in 2040 due to the Official Energy Pathways (shaded bars) compared to the Current Reduction Plans (white bars).

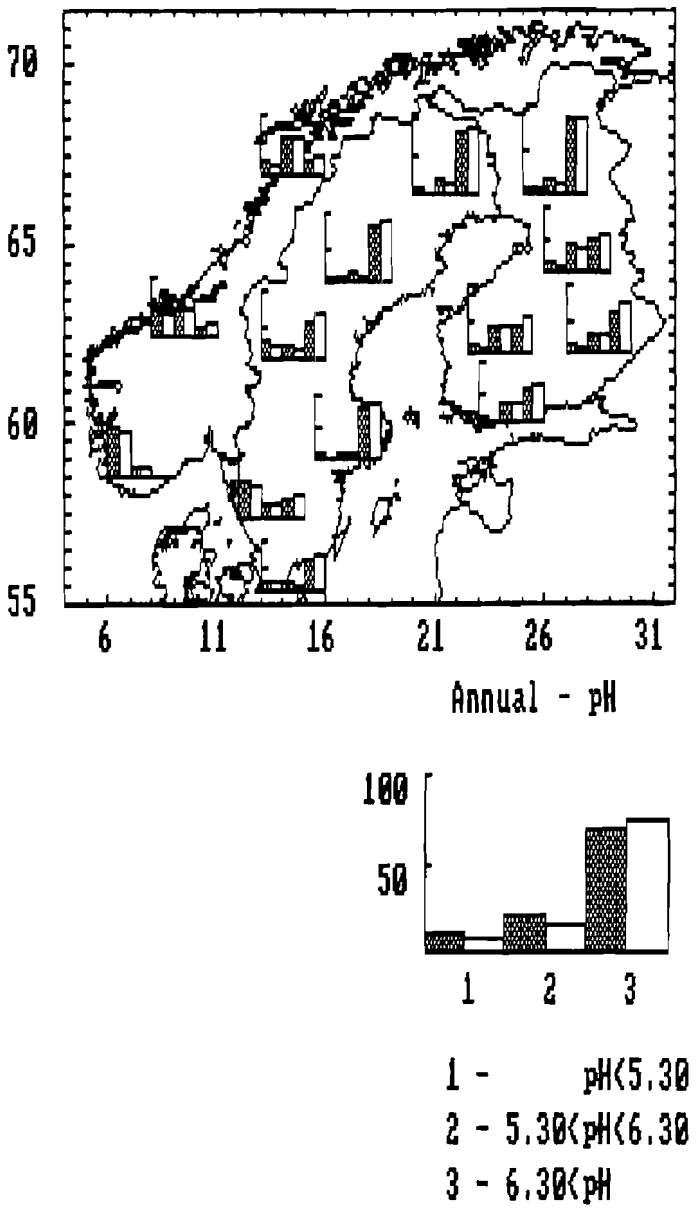

Figure 2.11 State of lake acidification in 14 predefined lake regions in 2000 due to the Official Energy Pathways (shaded bars) compared to the Maximum Feasible Reductions (white bars)

However, whether a lake is sensitive or not depends on many factors, e.g. soil characteristics, precipitation levels, lake depth, catchment areas and the location of lakes (e.g., surrounded by a forest). All these characteristics combined with particular deposition levels lead to differences in the state of lake acidity in a region. The question treated in this study is whether the application of a calibration procedure in a fixed zone allows for model predictions with a sufficient confidence resolution. In chapter 4 it will be shown that this is not generally true.

Therefore, the success by which a policy is defined that applies to many kinds of lakes, rather than an aggregate, is dependent on the extent to which the system of watershed acidification has been assessed. The assessment, i.e. the application of a model, should assure consistency between the calibration conditions and the conditions for which predictions are to be made. This study introduces a concept of model use, that relaxes both the calibration effects and the model application to predefined 'lake regions'. The concept draws for an important part on uncertainty analysis. Therefore much attention is paid to uncertainty analysis of the lake model (Kämäri et al., 1986; Gardner et al., 1987; Hettelingh and Gardner, 1988; Hettelingh et al., 1988) in this study.

Uncertainty analysis in RAINS

Besides the importance attributed to uncertainty in the lake module mentioned above, also in other modules of RAINS, attention is given to uncertainty.

Uncertainty analysis has been applied to the results of the RAINS deposition module (Alcamo and Bartnicki, 1987) by means of the original atmospheric transport model (Saltbones and Eliassen, 1983), from which the RAINS source receptor matrix was derived. The analysis aims at describing the confidence limits of the predicted deposition patterns over Europe. The results of the analysis is described in Alcamo and Bartnicki (1987), and applied by means of the computation and plot of a confidence interval around deposition patterns. In Figure 2.12 the light shaded area depicts the confidence range around a 4 $g/m^2/yr$

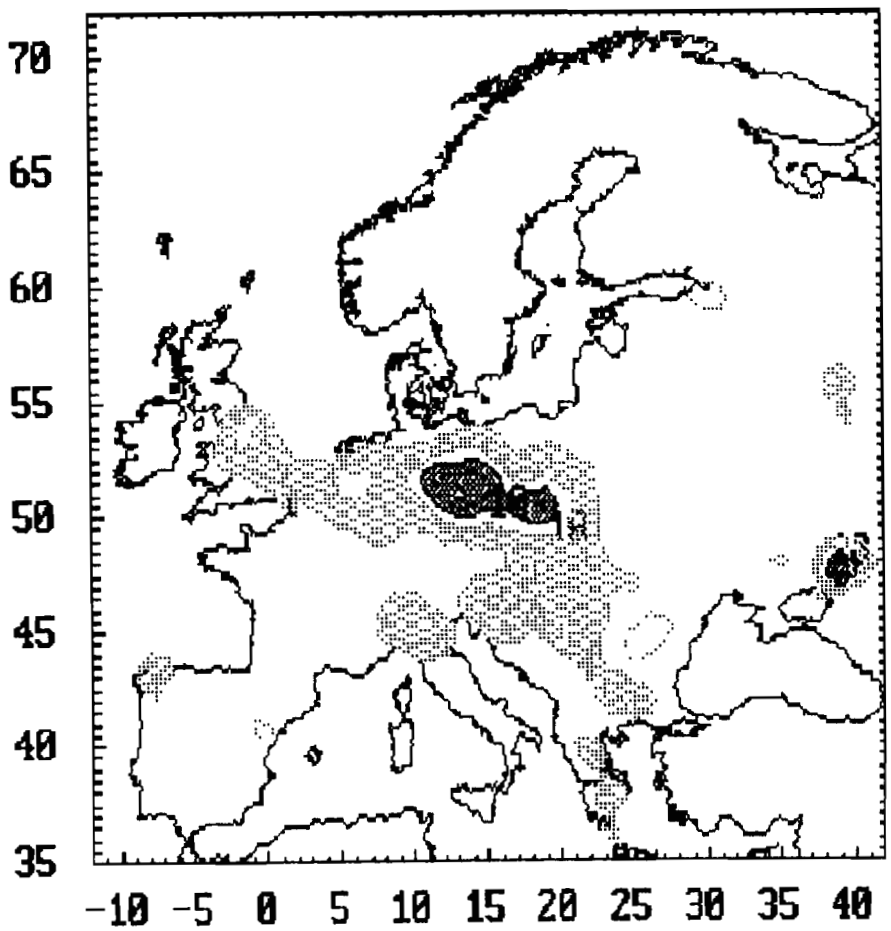

Figure 2.12 Deposition isoline of 4 g/m^2/yr including an upper and lower uncertainty range, depicted by the light shaded area. The dark shaded area represents the deposition of 10 g/m^2/yr including its uncertainty.

deposition isoline over Europe in 1995, whereas the darker shadings refer to the range around 10 $g/m^2/yr$ isolines in that year.

The effects of optimal emission abatement strategies as function of uncertain deposition patterns were investigated in the energy-emissions computation routines (Amann, 1989; Batterman and Amann, 1989). It was shown that reallocation over European countries of funds to finance sulfur abatement could lead to a reduction of the average costs to obtain targeted deposition levels in Europe.

Variables most sensitive for the behavior of the soil acidification module were depicted by Posch et al. (1985).

Uncertainty in IEMs in general

In general, the role of uncertainty analysis and the related use of stochastic techniques have received rather little attention in the development of IEMs. Brouwer (1987, p. 129) mentions, in relation to his treatment of statistical and econometric tools to operationalize an IEM, that "The lack of reliable information would lead to skepticism regarding numerically quantified modeling results. When either theoretical knowledge concerning the specification of an IEM is scarce and/or when relevant information is not reliable or not available at all, graph theoretic methods and qualitative calculus are useful tools to analyze the structure of the impacts between variables or to solve a set of equations in a qualitative way". Brouwer's (1987) treatment of qualitative and quantitative modeling methods is not extended towards the use of stochastic simulation to solve complex systems of equations and the related concept of uncertainty analysis.

The lacking treatment of uncertainty analysis in integrated modeling is acknowledged by Braat en van Lierop (1987b, p.67) who, with respect to uncertainty, note that "Explicit studies which evaluate integrated modeling in this respect have not been found among the studies surveyed".

2.3 UNCERTAINTY ANALYSIS

Errors in data measurements, model structure, model parameter estimation and numerical computation methods are a few of the causes that affect confidence in model predictions.

O'Neill (1971) appears to have been the first to directly address the contribution of individual parameter errors to the uncertainty in model results, using a Monte Carlo approach. Since then the term uncertainty, error or sensitivity analysis, applied to systems of the natural environment, can be encountered in literature on water quality modeling (Beck and van Straten, 1983), long range air pollutant transport (see also Alcamo and Bartnicki, 1987), dispersion of radionuclides (see also Helton and Iman, 1982; Helton et al., 1985; BIOMOVS, 1988), water acidification (Hornberger and Cosby, 1985a; Hornberger et al., 1986; Kämäri et al., 1986) , and ecosystems (see also Gardner, 1984; Gardner et al., 1980a, 1980b, 1982, 1987 ; Gardner and O'Neill, 1981, 1983; O'Neill and Rust, 1979; O'Neill and Gardner, 1979; O'Neill et al., 1980, 1982; Goldstein and Ricci, 1981; Bartell et al.,1983, 1986, 1988; Wood, 1985).

There does not seem to be a consistent terminology in the literature especially with respect to the distinction between uncertainty and sensitivity analysis. Tomovic and Vukobratovic (1972) state that "Sensitivity analysis studies the effects of parameter variations on the behavior of dynamic systems". This definition is sometimes referred to as conventional sensitivity analysis (Beck, 1987, p. 1422). In conventional sensitivity analysis the approach is based on the partial derivatives of the model structure with respect to individual model parameters. The techniques used in conventional sensitivity analysis assume that measurement errors do not change the patterns shown by the partial derivatives (Gardner et al., 1980a). With the introduction of Monte Carlo Methods the need for an infinitesimal approach to parameter changes is relaxed. In this context the sensitivity of model predictions may be estimated by setting the variance of all parameters to 1% of their nominal values (Gardner, 1984).

Uncertainty and error analysis are used interchangeably in the literature. The term error analysis , however, is often used in numerical techniques as applied to the accuracy of a function (e.g. Runge-Kutta). In uncertainty analysis the influence of the uncertainty,

i.e the variance, of a parameter on the model response is investigated. The relation between sensitivity and uncertainty analysis is formulated in the following mathematical representation, borrowed from Janssen et al. (1988).

Let **y** be a response defined by a time dependent relation of the model parameters[6] x_1 .. x_k as follows:

$$y = f(x_1, x_2, \ldots, x_k, t) \qquad (2.1)$$

For the sake of notational simplicity, equation 2.1 does not include state variables and input and output disturbances. Assume further that each parameter is fixed at a pre-specified real number value. Then the variation with respect to the values of x_i (i=1,..,k) leads to the following distortion of the value of y:

$$dy/dx_i = df(x_1, x_2, .., x_k)/d(x_i) \quad (i=1,..,k) \qquad (2.2)$$

which in conventional sensitivity analysis is used as a measure for the sensitivity of model responses.
Next, define the absolute deviation of the real value of a parameter x_i from its approximation x_{i0} by means of the mathematical operator **D** as follows:

$$Dx_i = x_{i0} - x_i \qquad (2.3)$$

Then, by applying **D** on **y**, the absolute deviation of **y** can be written as (Abramowitz and Stegun, 1964, p. 14, 3.5.5):

$$Dy = (df/dx_1)\, Dx_1 + (df/dx_2)\, Dx_2 + \ldots + (df/dx_k)\, Dx_k \qquad (2.4)$$

Assume further that $x_1, \ldots, x_k$ are uncorrelated so that:

$$E(Dy)^2 = (df/dx_1)^2\, E(Dx_1)^2 + \ldots + (df/dx_k)^2\, E(Dx_k)^2 \qquad (2.5)$$

---

[6] parameters have unique values during the course of a single model run, although these values may be varied from one run to the next. In this study, therefore model variables are denoted as parameters when there values are sampled once and remain the same during one Monte Carlo run.

where E is the mathematical expectation operator.
When investigating the influence of parameter distortion around a real value a logical next step is to use the mean as parameter estimator, changing equation 2.3 to:

$$Dx_i = \text{mean}(x_{i0}) - x_i \qquad (2.6)$$

From equation 2.5 and 2.6 now follows,

$$\text{var}(y) = (df/dx_1)^2\ \text{var}(x_1) + \ldots + (df/dx_k)^2\ \text{var}(x_k) \qquad (2.7)$$

where,
var(y) = the variance of y

Equation 2.7 shows that the contribution to the uncertainty, i.e. variance, of y on the left hand side of 2.7 is due to the uncertainty of x_i, i.e $\text{var}(x_i)$, and to the sensitivity, df/dx_i, of x_i, (i = 1 ... k) on the right hand side of 2.7. The result is that very sensitive parameters with low uncertainties, may still lead to a relatively low uncertainty of model response.

The derivation 2.1 to 2.7 formally only holds if a Taylor expansion in 2.4 is allowed. This is only the case for small deviations resulting from the operator D.

Distinction of errors in the modeling process.

Modeling a system is error prone in several ways. Beck (1987, p.1396) developed a frame of reference (Figure 2.13) of uncertainty, which he defines as follows:

"...uncertainty will be understood in a probabilistic sense, encapsulating therefore the variability in the outcome of a random event (including the attributes of a biological population) and embracing the notions of erroneous assumptions or the distributions of errors associated with observed or estimated quantities" (Beck, 1987, pp. 1395-1396).

The explicit introduction of stochasticity in the concept of

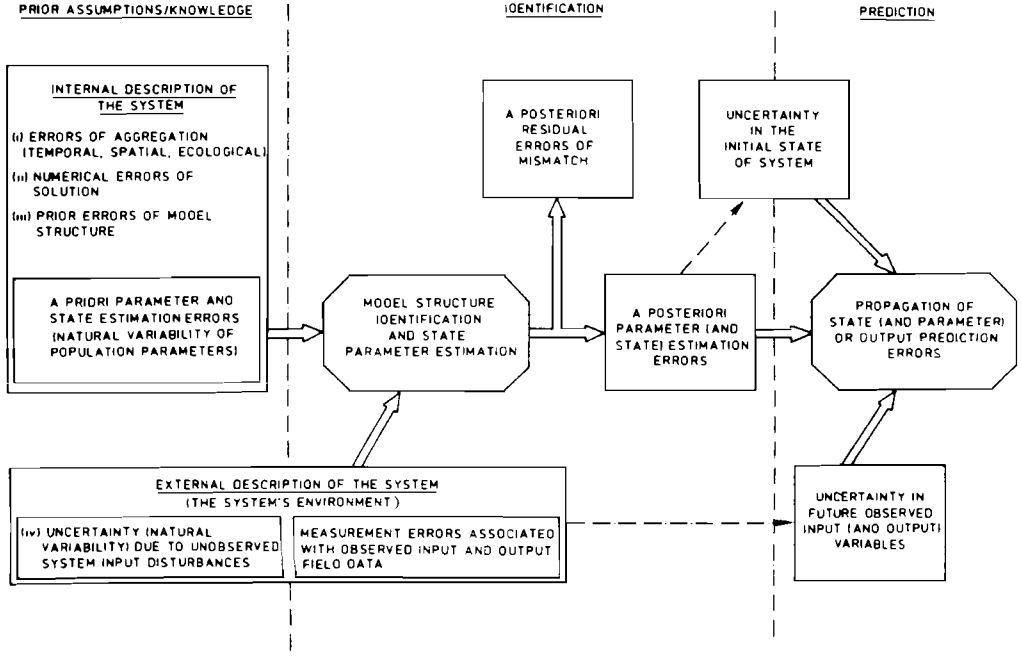

Figure 2.13 Frame of reference for the analysis of uncertainty (Source: Beck, 1988, p. 1396)

uncertainty overlaps with the definition of Mihram (1972) presented in chapter 1 of this study. One of the causes of uncertainty in the prior assumptions about the system, as shown in Figure 2.13, is directly related to the problem of aggregation.

The essence of Beck's treatment of aggregation with respect to water quality modeling is that " ..detailed spatial patterns of water circulation and equally detailed differentiation of ecological behavior described by the more complex models would demand experimental observations that are simply not technically feasible" (Beck, 1987, p. 1396). Aggregation is on the one hand related to the temporal, spatial and ecological scale of the system, and on the other to the number of system compartments distinguished in the model structure. Beck argues further that the debate about the approaches to modeling can be shifted away from the usual confrontations between the statistical and the mechanistic models. This confrontation may be relaxed by the notion that complex models may serve the understanding of a system, whereas simpler, statistical, models are more suited for the stage of model analysis (see section 2.1). Statistical models, sometimes referred to as empirical models, comprise response surface methodologies and metamodeling whereas mechanistic models (see also Reckhow and Chapra, 1983, pp. 20) represent the more reductionist view of systems (see chapter 1). Several studies have demonstrated the success of such aggregation. O'Neill and Rust (1979), for example, showed that non linear models with two interacting components could successfully be substituted by a single variable model. Cosby et al. (1985b) demonstrated that long-term catchment responses to acidic depositions, can be represented by a simplified representation of the different processes. Gardner et al. (1982) showed that a complex non linear model and a relatively simple empirical model could both be used to represent the same set of system dynamics. A similar result has been obtained by Hettelingh and Gardner (1988) with respect to the RAINS lake model (see chapter 4 of this study). In this study the term aggregation will have a twofold meaning:

1. the aggregation of a model structure, to sufficiently describe the process of acidification of a set of lakes in a geographic region, and
2. the notion that a geographic region of lakes is in fact an aggregation or, more specifically, a unification of many

different kinds of lakes to one or more classes of lakes with similar characteristics.

The aggregation of a system to compartments is an exercise that also lies at the roots of the model structures discussed earlier, i.e. the vertical, horizontal, satellite and multi-layer structure. Both the IEM structure and the ecological model structures, referred to above, have in common that several mechanisms are integrated. In an IEM the mechanisms may be related to economic and other 'macro' processes, whereas in an ecological model of for example water-systems, more detailed 'micro' processes, like water percolation in soils, govern the mechanisms. The influence of the model structure on uncertainty is however not only restricted to the level of systems representation, i.e. macro or micro, but also to the nature of the equations. Model structure uncertainty is extremely difficult to quantify in a formal manner, such that it can be distinguished from other sources of uncertainty. The sources of uncertainty thus often accounted for are uncertainties in the initial state of the system, uncertainty in model parameter estimates, uncertainty in the observed input disturbances and output responses and uncertainties due to unobserved input disturbances of the system (Beck, 1987, p.1396; see also O'Neill and Gardner, 1979).

The major part of methods used to investigate these uncertainties, are based on random sampling methods that are very often referred to as Monte Carlo. The distinctions made in chapter 1 between Monte Carlo methods, Monte Carlo simulation and stochastic simulation are very scarcely elaborated in the reported applications of uncertainty analysis. Other methods, mentioned in the literature applying uncertainty analysis, are the already mentioned (chapter 1) analytical approach, the Fourier Amplitude Sensitivity Test (FAST) (see also Cukier et al., 1978; McRae et al., 1982; Liepmann and Stephanopoulos, 1985; Uliasz, 1985), and methods based on possibility theory (see also Keesman and van Straten, 1987). In the FAST method output variables of a model are Fourier analyzed. The Fourier coefficients represent an average of the output variables over the uncertainties (variations) of all the parameters (Cukier et al., 1978, p.4). In the methods based on possibility theory model parameters leading to a desired model behavior are chosen on the basis of a definition of a membership function. Keesman and van Straten (1987) apply possibility theory in combination

with Monte Carlo analysis, which method is referred to as the Modified Monte Carlo-Set Theoretic Method, to reflect model as well as data uncertainty in model predictions. The method assumes that there is a gradually diminishing possibility to find the true system behavior further away from an observed data point, which allows for the definition of membership function values to each data-point. Next, a criterion function may be defined which can be optimized. A parameter vector can for example be chosen that maximizes the lowest membership function value over the time-series of data-points. The resulting model response is finally compared to predefined system bounds (see Keesman and van Straten, 1987, pp. 299-301).

An overview of uncertainty methods can be found in Janssen et al. (1988) . In the vast majority of applications in environmental modeling, sensitivity and uncertainty analysis is related to Monte Carlo methods.

Before presenting a more formal description of sampling methods used in Monte Carlo simulation, first its application in the stage of model synthesis and model verification is treated in the next section. This part of model identification is often referred to as calibration.

2.4 CALIBRATION AND MONTE CARLO SIMULATION

The aim of calibration is to test a model structure, representing a given system, under uncertainty. This method may involve the recursive adaptation of model parameters until the comparison between the model outputs and a set of numbers, like measurements, fulfills a predefined criterion (See also Fedra, 1983), e.g. a non-significant difference between the cumulative distribution of model predictions and measurements. Kleijnen (1987, p.337) restricts the term calibration to the following methodology: model parameters are quantified from, say, N_1 number of runs with the model, whereas, say, N_2 number of other runs are executed to validate (see section 2.1) the calibrated model. Beck (1983, 1987) extends the concept of calibration to the investigation of the appropriateness of a model structure to be capable of describing measured system's behavior.

The methodologies of calibration, discussed in the literature, have in common that they are based on Monte Carlo simulation and/or that a recursive procedure is involved. In the development of the ecological module of the GMM, for example, the appropriateness of model parameters was solely recursively adapted until a reasonable, not statistically tested, representation of the real world was obtained.

No classification of calibration methods in environmental modeling has been encountered in the literature. It appears that most of the concepts encountered, are based on an approach applied in research by Young (1978), Hornberger and Spear (1980;1981), Spear and Hornberger (1980) and Young (1983), that is aimed at analyzing model uncertainty, system identification and model prediction. This method, often called regionalized sensitivity analysis which approach is denoted by Beck (1987) as the Hornberger-Spear-Young (HSY) algorithm, and the resulting approaches that are based on the HSY algorithm are discussed below.

Regionalized sensitivity analysis.

The purpose of regionalized sensitivity analysis is to generate a set of model parameter vectors from repeated samples that leads to a particular model response, that reflects the observed system behavior, further referred to as the desired model behavior **B**. Let the model response that does not reflect the observed system behavior be **non-B**. For this purpose Monte Carlo simulation is applied to the model. Each run uses another sample of the model parameters. The result of the method is a set of parameters that gives rise to the desired model behavior (**B**) and a complementary set (**non-B**) of parameters that does not (Figure 2.14). The mathematical formulation presented below is taken from Young (1983, pp.75-76). This formulation is chosen to be able to more specifically describe the method of regional sensitivity analysis in this study. The method can however also be applied to other classes of functions and with discrete time data.

Consider a representation of a general class of systems by a non-linear, state space differential equation in continuous time, as presented in equation 2.8

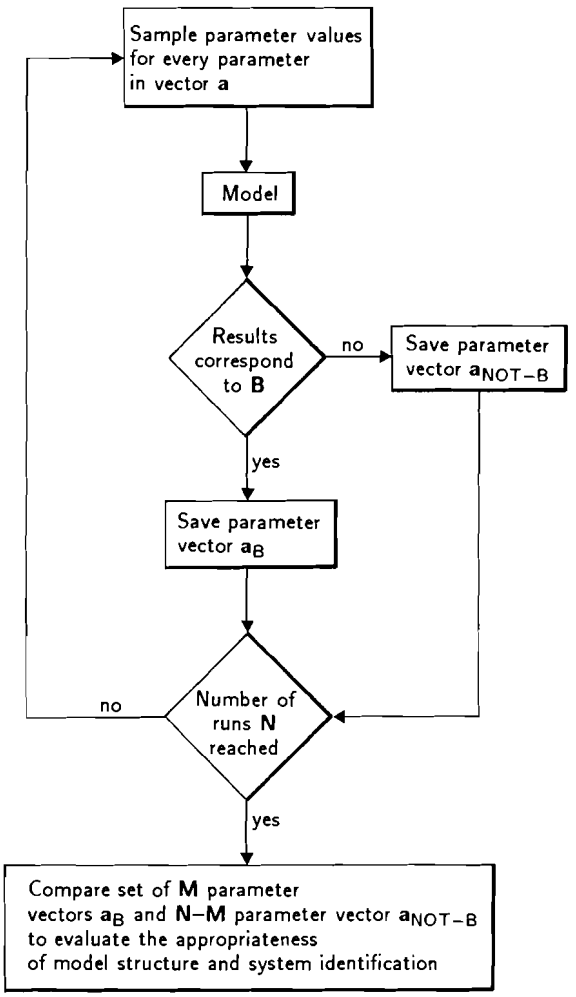

Figure 2.14 Hornberger-Spear-Young algorithm

$$d\mathbf{x}/dt = \mathbf{f}_i(\mathbf{x},\mathbf{a},\mathbf{p}^c,\mathbf{p}^d,\mathbf{U},t) \qquad (2.8)$$

where,

t = time

$\mathbf{x}$ = $[x_1, x_2, \ldots, x_n]^T$ is an n-dimensional vector of state variables which describe the system behavior in the "state space".

$\mathbf{a}$ = $[a_1, a_2, \ldots, a_q]$ is a q-dimensional vector of (possibly time-dependent) parameters or coefficients that characterize the system in the state space.

$\mathbf{p}^c = [p_1, p_2, \ldots, p_m]^T$ is an m-dimensional "control input" vector.

$\mathbf{p}^d = [d_1, d_2, \ldots, d_l]^T$ is an l-dimensional vector of measurable (exogenous) but uncontrollable disturbances which affect the system

$\mathbf{U} = [u_1, u_2, \ldots, u_n]^T$ is an n-dimensional vector of stochastic disturbances whose statistical properties may or may not be known, depending on the level of a priori information available about the system.

$\mathbf{f}$ = a nominally nonlinear and non-stationary vector function reflecting the idea that badly defined systems will, in general, exhibit nonlinear and possibly changing behavioral patterns. Since many physical relationships are stated more naturally in continuous time, a continuous time formulation has been chosen.

The approach consists of evaluating the model behavior for different vector functions $\mathbf{f}$ with the associated parameter vector $\mathbf{a}$ which is represented as the probability distribution over the complete range of, supposedly, possible values of the coefficients that compose the vector. The approach should also allow for the input uncertainty, $\mathbf{U}$, to represent disturbances to the system. In addition it may be desirable to consider the model behavior for different deterministic inputs $\mathbf{p}^c$ and $\mathbf{p}^d$, to simulate particular system conditions, e.g. "dry" or "average" precipitation conditions. This approach is exploited in methodological terms by recourse to Monte Carlo simulation analysis, in which equation 2.8 is solved repeatedly for uncertain parameters and inputs by sampling at random from their assumed parent distributions. The result of the analysis is a large number of random results, each providing a unique state trajectory $\mathbf{x}(t)$. Finally the test is made whether the sampled model parameter vector, $\mathbf{a}$, leads to a state

trajectory **x**(t) that gives rise to an expected behavior **B** or to the complement of the behavior set **non-B**. The number of Monte Carlo runs will thus consist of **N** runs in which **M** parameter vectors lead to the behavior **B** and **N-M** did not. In evaluating which elements of the vector **a** are giving rise to the behavior **B** and which did not, the cumulative probabilities associated with these elements resulting from **M** samples are compared to those resulting from the **N-M** samples. An element of vector **a** is considered important if there is a statistically significant difference between the cumulative distributions based on resp. **M** and **N-M** samples, and the vector **a** is considered not important otherwise. A case study based on this principle for distinguishing between the parameters that significantly influence model response and those that do not, can for example be found in Hornberger et al. (1986). The statistical tools used may be the Kolmogorov-Smirnov, or the Mann-Whitney test or the tools may be based on eigenvalue-eigenvector analysis of the variance-covariance matrices associated with the parameter vectors by means of principal-component methods (Young, 1983, p.77).

The philosophy of the HSY algorithm has been applied frequently for model calibration and for obtaining model predictions, although the actually applied methodology varies between the authors. The application may for example be aimed at using the set of parameter-vectors, that resulted in behavior **B**, for model prediction.

O'Neill et al. (1980), for example, applied Monte Carlo simulation to a simple model that represents the dynamics of herbivores, such that only those simulation results were kept that led the model to pass through 4 "windows", i.e. model response constraints, in time.
In this application, the aim was to investigate parameter errors, for the entire time series for the model simulations that matched the "window" criterium. The difference with the genuine HSY algorithm is that sampled parameter values are varied to investigate prediction error, and not so much aimed at selecting the right parameters.

In the calibration of the RAINS Lake Module two approaches have been used. The methods are generally applicable and are not dependent on the model equations, which in the case of the RAINS Lake Module are presented in chapter 3. The first method is implemented in the current RAINS model, to be able to make predictions about lake acidification in 14 a priori determined lake regions in northern Europe. The method (see

Kämäri, 1988; Posch and Kämäri, 1987) is depicted in Figure 2.15. The approach is aimed at saving the sampled values of only the model parameters, that lead to model predictions of pH and alkalinity of which the joint frequency distribution matches the joint frequency distribution of the field measurements of pH and alkalinity. Otherwise, the set of parameter values is discarded. In fact the saved set of parameter values, in the terminology of the HSY algorithm, lead to a behavior of model simulations such that the predictions match the measured pH and alkalinity distribution. This 'filtering' procedure (Figure 2.15) may mathematically be described as follows:
Let,

$$g(ph,alk)_i \tag{2.9}$$

be the frequency of measurements in the two dimensional-interval[7] (i.e. a square) **i** of the range of measured pH and alkalinity combinations. Let further,

$$s(a,SpH,Salk)_j \tag{2.10}$$

be the joint frequency distribution of a combination of pH model response values (**SpH**) and alkalinity model response values (**Salk**) that fall in the two-dimensional interval **j**, for a particular set of parameter-values of the parameter vector **a**. Note, as mentioned before, that the term parameter is used for model variables of which the values have been sampled and thus remain the same during one single run.

State variables are left out of the equations 2.10 and the following equations for notational simplicity. For each set of parameter values **a** that lead to a model output falling in **j**, a value, e[8], is added to equation 2.10 as follows:

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[7] **i** is a notational simplification of the square defined by the lower left hand corner $(ph^l, alk^l)_i$ and the upper right hand corner $(ph^r, alk^r)_i$.

[8] **e** should be larger than the inverse of the number of Monte Carlo runs.

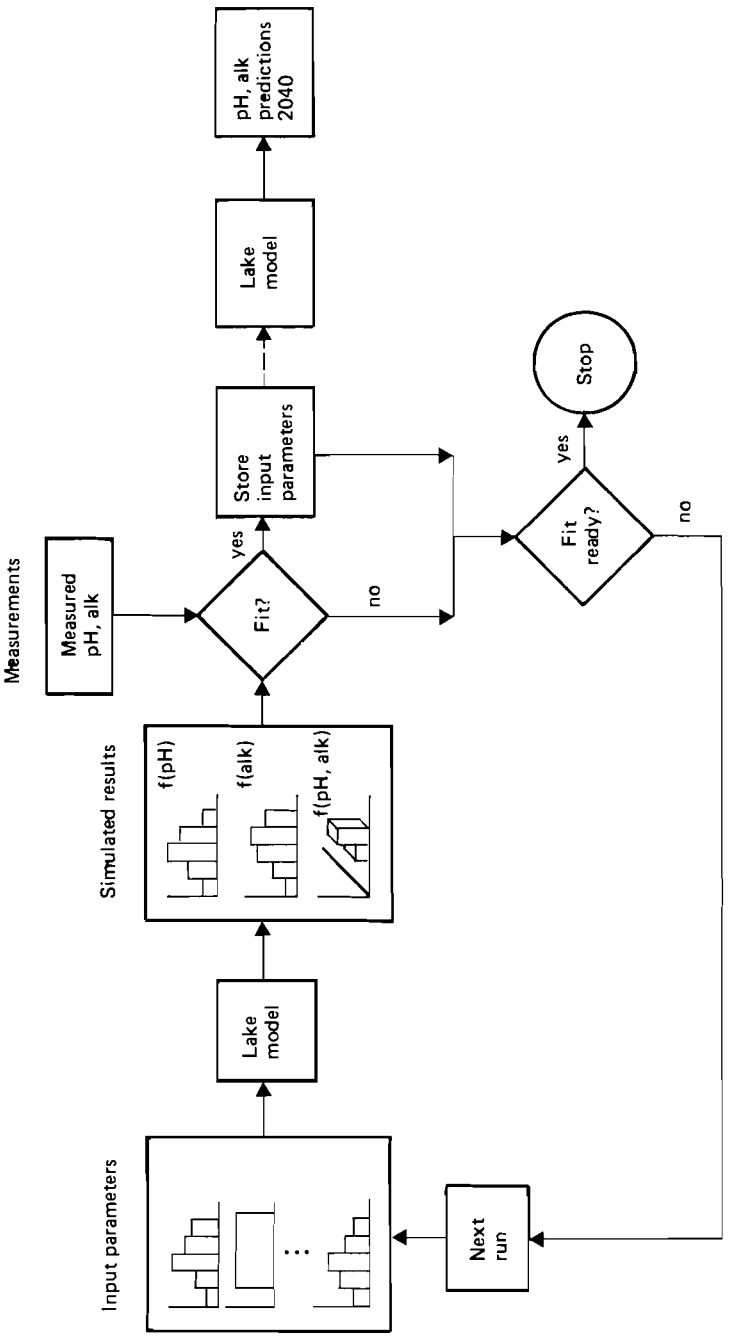

Figure 2.15 Filter procedure for calibration of the RAINS Lake Module leading to parameter values used for the prediction of lake acidification.

$$s(a_k, SpH_k, Salk_k)_j = s(a_1, SpH_1, Salk_1)_j + e \qquad (2.11)$$

where,

k,l = Monte Carlo run number k and number l,

$1 < k < N$ = total number of simulations

The parameter values a_n, with n denoting one of the M simulations $[n_1, n_2, \ldots, k, l, \ldots n_M]$, with $M < N$, leading to a model output in two-dimensional interval j, are saved, provided that

$$s(a_n, SpH_n, Salk_n)_j \leq g(pH, alk)_i \quad \text{for } i = j \qquad (2.12)$$

This procedure is repeated for all i in the allowable range of measured pH and alkalinity values, until s becomes equal to g in 2.12 for all j. The set of parameter values of all, say M, runs which does not lead to the violation of 2.12 is saved. The set that lead to model predictions violating 2.12, i.e. lead to predictions that are outside of the allowable range of measured pH and alkalinity values, is rejected. Finally, the M sets of accepted parameter values are used to predict the distribution of lake acidity in future years as a result of forecasted sulfur deposition. The computational disadvantage of this approach is (1) that the important as well as the unimportant parameters have to be manipulated, and (2) that the number of Monte Carlo runs needed cannot be fixed a priori; it may very well happen that after for example 1000 runs some ranges of the measured pH and alkalinity frequency distribution have not been reproduced in the M accepted pH and alkalinity model predictions (M<1000). A more efficient algorithm taking care of the latter disadvantage has been developed by Posch (1987) and is added in appendix I of this study.

The second method applied to the RAINS lake module was developed by Gardner et al. (1987) and Hettelingh and Gardner (1988) circumvents the first disadvantage of the filtering method mentioned above. This method incorporates the influence of parameter uncertainties and will be further discussed in chapter 4 of this study.

Monte Carlo Simulation in sensitivity and uncertainty analysis

With respect to the use of Monte Carlo methods for the purpose of

sensitivity and uncertainty analysis on computer models in general and environmental models in particular modeling, much recent progress, discussed earlier, is due to the work of Iman and Conover (1980), Iman et al. (1981) and to McKay and Beckman (1979).

In the application of Monte Carlo simulation in the context of sensitivity and uncertainty analysis the following two concepts are of importance,

1. the sampling method:
   Sampling from parameter value ranges can basically be done in three ways, i.e. random sampling, stratified sampling and Latin Hypercube sampling.
2. the analysis of the relative importance of model parameters: the issue here is to quantify how much each of the parameters is contributing to the model response.

Sampling methods

Adapting equation 2.1 to represent model output as function of time, the next equation represents the output **Y** by the computer code as a result of any selection of input parameters $\mathbf{X} = (X_1, X_2, \ldots, X_k)$. Note, as before, that the term 'parameters' is used rather than 'variables', because every Monte Carlo run will constitute a sampled value of **X** that remains constant in that particular run, even though this run may contain several time steps.

$$\mathbf{Y(t) = f(X)} \qquad (2.13)$$

The uncertainty about the values of the input parameters is modeled by treating them as stochasts, and is obtained by studying the probability distribution of **Y(t)**. Assume that the inputs **X** have a known probability distribution **F(X)**, for **X** defined in a sample space **S**. McKay and Beckman (1979) distinguishes three methods by which input values $\mathbf{X}_1, \mathbf{X}_2, \ldots, \mathbf{X}_n$ can be obtained:

1. Random Sampling. This consists of a straightforward drawing of $\mathbf{X}_1, \mathbf{X}_2, \ldots, \mathbf{X}_n$ from **F(X)**, in a not necessarily more complicated way than repeatedly tossing a dice for the case where the value range of

the stochast lies between 1 and 6.

2. Stratified Sampling. In stratified sampling, all areas of the sample space of X are represented by input values. Let the sample space S of X be partitioned in I disjoint strata, with not necessarily the same size, i.e. $p_i = P(X \subset S_i)$. A random sample X_{ij}, j=1, ... ,n_i where n_i will sum to N for all i of I. In other words, N runs will consist of n_1 hits in stratum 1, n_2 hits in stratum 2, ... , n_I hits in stratum i. The number of samples from a stratum S_i depends on its size p_i. If, I = 1, then random sampling is applied over the entire sample space.

3. Latin Hypercube Sampling. With this method the aim is to ensure that each of the input parameters X_k, k = 1, ... ,K, has all portions of its distribution represented by input values. Here, in other words, sampling is done without replacement, i.e. to sample all portions of S. The procedure consists of dividing the range of each X_k into N strata of equal marginal probability 1/N, and sampling once from each stratum. One sample is denoted by X_{kj}, j = 1, ... ,N, k = 1, ... ,K and called the X_k component of X_i, i = 1, ... ,N. Note that the N intervals of each component combine to form N^k cells which cover the entire sample space of X. All components are combined in a random manner.

Much attention has been spent to the relative advantages of the sampling methods presented above. Generally, with respect to Monte Carlo simulation, latin hypercube sampling has the following advantages:

1. Variance reduction: The variance of the sample mean obtained with Latin Hypercube is smaller or equal to the variance of the sample mean resulting from other sampling procedure, provided that Y(t) is monotonic in each of its arguments. The proof of this statement can be found in McKay and Beckman (1979). Stein (1987) shows that the asymptotic variance of an estimator based on Latin Hypercube sampling is not only smaller than in sample random sampling, but that in addition the amount of variance reduction increases with the degree of additivity in the random quantities.

2. Representative sampling: Even with a relatively small sample, the method assures that values are drawn over the entire probability range

of a parameter.

3. Parameter correlations: By implementing a method developed by Iman and Conover (1982), a desired rank correlation matrix may be induced on the multi-variate input random parameter **X**, defined above. The random pairing of intervals by Latin Hypercube may similarly lead to undesired pair-wise correlations among some of the parameters, especially when the sample size is small. The addressed method can avoid this. This addition to latin hypercube sampling is called Modified Latin Hypercube sampling (see Iman and Helton, 1988).

Computer programs have been written that allow for an easy implementation of any model in a Monte Carlo framework using latin hypercube sampling or its modified equivalent. For this study the program written by Gardner et al. (1983) has been implemented on the RAINS Lake Model. The program, called PRISM, will be treated in some detail in chapter 4.

Relative importance of parameters

The purpose is to analyze which of the input parameters contributes most to the uncertainty, i.e. the variance, of the model response. Three methods are used:

1. Regression techniques: Ordinary Least Squares is applied treating the model response as dependent variable, and the input parameters as the exogenous variables. Thus the computer model is in fact replaced by a single equation model response as a function of input parameters, for which Kleijnen (1987, pp. 147-157) introduces the term metamodel. In other literature (see also Downing et al., 1985) the term response surface is encountered more frequently. By investigating the coefficients of the parameters an, optionally stepwise, evaluation is obtained of the importance of each parameter (see also Iman and Conover, 1988). The relevance of the linear approximation can be obtained by inspecting the R^2 (see also Gardner et al., 1987) and the relevance of parameter X_k is investigated by its contribution to the R^2 statistic. This partial R^2 is computed by

$$PR2_k = (RSS - RSS_k)/TSS \qquad (2.16)$$

where,

$PR2_k$ = the partial R^2 for parameter X_k

RSS = Regression sum of squares for the complete model

RSS_k = Regression sum of squares for the reduced model, i.e omitting parameter X_k

TSS = Total sum of squares

If the parameters are nearly orthogonal, $PR2_k$ represents the fractional contribution of X_k to the variance of the model output Y, and is therefore an important indicator in uncertainty analysis. This method has been incorporated in many applications of PRISM (Gardner et al., 1983).

The use of response surface techniques may be aimed at replacing the full computer model, in order to be able, for example, to make predictions about a system for which purpose the full model may be too complex. All inferences about the uncertainty and sensitivity analysis may be derived from this fitted equation only. A careful consideration is necessary of the structure of the metamodel and the parameters to include; questions about the inclusion of quadratic and cross terms in the equation may easily lead to a model equally difficult to handle. The fraction of the parameters to include can be evaluated by methods referred to as fractional factorial design (see also Kleijnen 1975).

Iman and Conover (1979;1981) point out that regression may lead to biased conclusions if the relation between model response and input parameters is nonlinear, which problem may be circumvented inter alia by applying rank regression. Rank regression consists of replacing the data with their corresponding ranks which is then used in an OLS procedure.

2. Correlation analysis: Computing correlation coefficients between the response variable and input parameters may also lead to insight in the parameter influences on the output variable. Either the Pearson or the Spearman correlation coefficient can be used. The Pearson correlation is applied to the parameter values. This method has the disadvantage that (see also Downing et al., 1985):

1. Extreme observations may have too much influence,

2. A measure of linearity is obtained, whereas a measure of monotonicity is desired to investigate non-linearity between X and Y.

The Spearman correlation coefficient, which computed using ranks in stead of values, relaxes both disadvantages of the Pearson measure of correlation. The partial rank correlation is defined as the correlation between the ranks of one parameter with the ranks of the response variable, removing the effect of the other parameters. When the partial rank correlation is close to 1, it indicates a strong monotonic effect of the parameter on the response. The disadvantage of using rank correlation is that only true sensitivities (e.g., between output and inputs) are obtained, and that relative sensitivities (e.g., the output is more sensitive to an input than to another) of parameters become difficult to perceive.

Generally both the partial correlation and partial rank correlation is computed. If the partial rank correlation is high but the partial correlation low, a nonlinear relation between response and input should be considered.

2.5 CONCLUDING REMARKS

An overview has been presented of the current state of integrated environmental modeling. In particular applications of uncertainty analysis in the field of ecosystem- and water quality modeling have been described. In these applications, models investigated are generally complex in nature, i.e. many, not necessarily linear relations are interacting in a way that does not allow for straightforward analytical solution methods.

The fact that depletion of our natural resources is due to many interacting different systems (and subsystems) has been described as a reason to favor modeling in an inter disciplinary rather than in a mono-disciplinary way. This has led to Integrated Environmental Modeling. Attention was given to the different levels of hierarchy and the different model-structures by which systems can be identified in integrated modeling, i.e. the level of detail and the linkages by which systems and subsystems can be represented to explain the same phenomena. This has led to the notion that there is similarity from a systems analysis point of view between integrated 'micro' processes, e.g. the modeling of chemical constituents in water quality modeling,

and integrated 'macro' processes, e.g. the functioning of groups of actors in a social economic system. Three examples of recent Integrated Environmental Models were presented, i.e. the Modeling and Information System for Environmental Policy, the Integrated Regional Environmental Model for Physical Planning and the Regional Acidification INformation and Simulation model. These three models were compared with respect to policy and user friendliness and the extent to which uncertainty and sensitivity analysis are included. It was shown that in Integrated Environmental Modeling in general uncertainty and sensitivity analysis is becoming increasingly important for the credibility of policy that is based on model-results.

Sensitivity and uncertainty analysis have been treated and associated with the apparent application of Monte Carlo simulation in the vast majority of applied uncertainty analysis in the field of environmental modeling.

Finally, different methods regarding Monte Carlo sampling and methods to investigate the contribution to the variance of model response due to model parameters, have been reviewed. A combination of latin hypercube sampling and metamodeling with related partial R^2 investigations between model inputs and outputs, emerged as a possible appropriate framework to investigate model uncertainties. A widely used computer program, PRISM (Gardner et al., 1983) incorporating these features will be used in this study.

This chapter and chapter 1 were concerned with the presentation of the state of the art of environmental modeling placed in a wide context of applied systems analysis. This context enabled the treatment of model aggregation and scales of model application. In the next chapters the context, just mentioned, will be treated in a much more focussed way. These chapters hence become very detailed and contain the equations of the RAINS Lake Model (chapter 3), the illustration of the possible insufficiency of calibration for the application of the model in fixed regions (chapter 4), and the presentation and illustration of an alternative zonal approach (chapter 5) followed by an application in chapter 6.

3. WATERSHED ACIDIFICATION MODELING

3.1 INTRODUCTION

Watershed acidification is the result of chemical reactions in the air, soil and watersheds involving sulfur and nitrogen oxides. Compared to 1960 the total emissions of SO2 in Europe have increased with about 66% and those of NO_x with about 144% in 1990 (RAINS data-base). The development of the total European emissions is shown in Figure 3.1.

The relationship between emissions of sulfur and nitrogen oxides and acidification, i.e. acid rain, is currently better understood since Oden (1968) first investigated acidification effects in the natural environment. The processes involved in the acidification of soils and watersheds are expressed in Figure 3.2. The representation of these processes in models have recently received much attention.

This chapter gives an overview of the subprocesses that are considered in many watershed acidification models to familiarize the reader with the concept. The scope of this study emphasizes the concept of applying model results to regions under uncertainty. It would be beyond the scope of this study to also provide the reader with a detailed description of the often complex chemistry involved in the processes. For this purpose, the reader is referred to Stumm and Morgan (1981). A verbal description of the processes will be presented instead, with the exception of the equations that are incorporated in the RAINS Lake Module. A comparison is made of three leading models with emphasis on the RAINS Lake Model with regard to process treatment and regionalization.

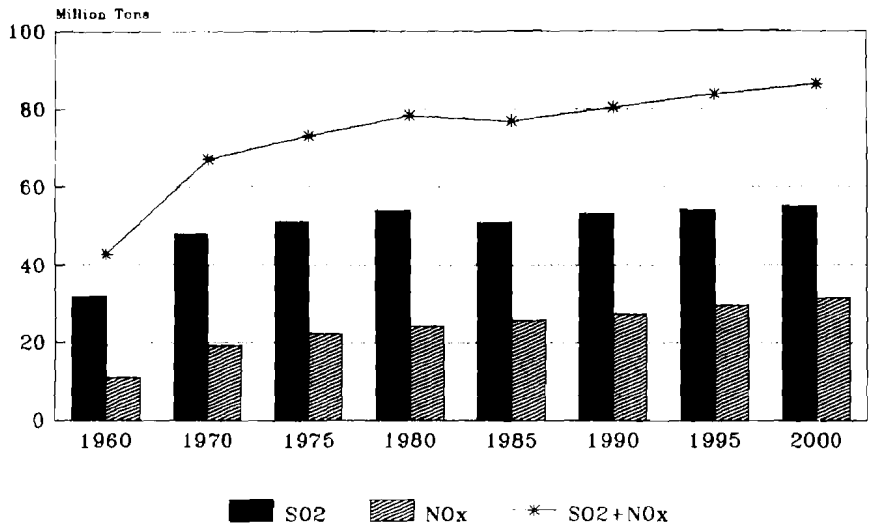

Figure 3.1 The time development of SO_2 and NO_x (units of NO_2) emissions in Europe in millions of tons. Emission forecasts are based on the official governmental energy use estimations until 2000 that have been implemented in RAINS.

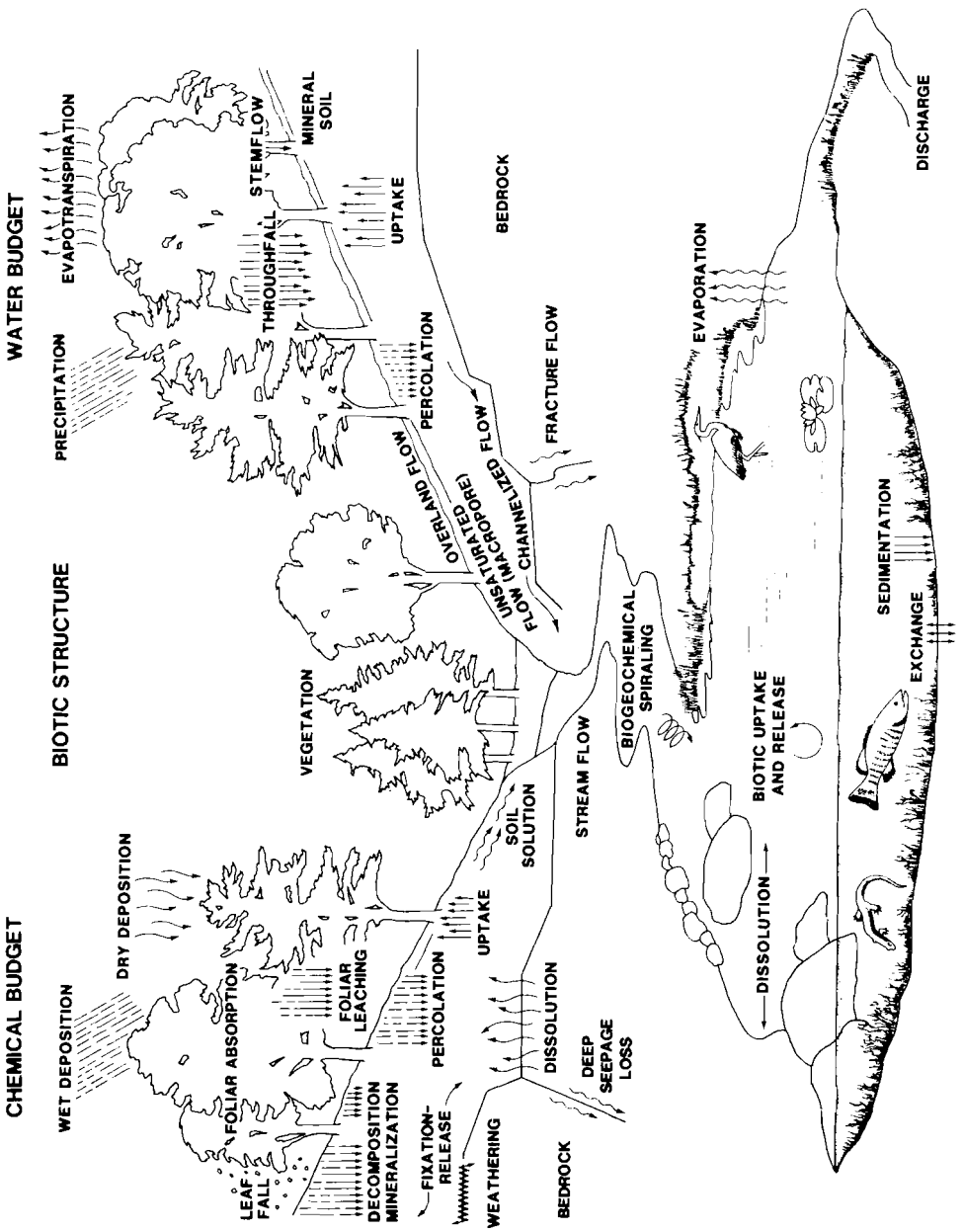

Figure 3.2 Processes considered relevant under the current state of scientific knowledge, to the contribution of watershed acidification (source: Cook, 1988, p. 20).

3.2 SUBPROCESSES IN WATERSHED ACIDIFICATION MODELS

As can be seen from Figure 3.2 many processes on the soil surface as well as in the soil precede the actual acidification of a lake. Many models have been developed dissimilar from one another with respect to the processes included (see Kämäri, 1988, p.17). An overview of the models is presented in Table 3.1.

Table 3.1 11 different watershed acidification models[1]

Model	Author(s)
Equilibrium	Reuss (1980), Reuss & Johnson (1985)
Birkenes	Christophersen et al. (1982)
Empirical	Wright and Henriksen (1983)
ILWAS	Chen et al.(1983), Gherini et al. (1985)
Adsorption Isotherm	Arp (1983)
ETD	Schnoor et al. (1984;1986)
PULSE	Bergström et al. (1985)
MAGIC	Cosby et al. (1985a;1985b)
Seepage Lake	Lin and Schnoor (1986)
Direct Distr. Model	Small and Sutton (1986)
RAINS Lake Module	Kämäri (1988)

[1] adapted from Kämäri, 1988, p.17

This study is not served with a detailed description of all of the models mentioned in Table 3.1. Three of the (American) models i.e. the Model of Acidification of Groundwater in Catchments (MAGIC) of Cosby et al. (1985a, 1985b), the Enhanced Trickle Down Model (ETD) of Schnoor et al. (1984, 1986) and the Integrated Lake/Watershed Acidification Study (ILWAS) of Chen et al. (1983) and Gherini et al. (1985) have received much attention not only in Europe but also by the National Acid Precipitation Assessment Program (NAPAP) in the United States (Rose, 1988). In the next sections of this chapter first an overview is presented of the chemistry involved in watershed acidification. Then the equations of the RAINS Lake Model are presented.

The basic chemistry

An acid is a substance that can donate a proton (i.e., H^+) to another substance, whereas a base is a substance that can accept a proton from another substance. A proton transfer can thus only take place if an acid reacts with a base. This reaction may be summarized as follows (Stumm and Morgan, 1981, p. 122)

$$\text{Acid}_1 = \text{Base}_1 + \text{proton} \quad (3.1)$$
$$\text{proton} + \text{Base}_2 = \text{Acid}_2 \quad (3.2)$$
$$\text{Acid}_1 + \text{Base}_2 = \text{Acid}_2 + \text{Base}_1 \quad (3.3)$$

The concentration of H^+, $[H^+]$, is measured in **pH** units defined as

$$\text{pH} = 3 - \log [H^+] \quad (3.4)$$

with $[H^+]$ expressed in eq/m^3.[9]

Acidification of natural waters is the result of interactions between acids and bases. Acids that naturally have leaked to watersheds, and the SO_x and NO_x contamination of precipitation on watersheds, react with bases that have been set free by the erosion of primary rock. This erosion process will further be called weathering. Depending on the weathering rate and the capacity of bases, called buffer capacity, the equilibrium process may lead to a changed concentration of H^+ protons.
The pH of natural waters generally lies between 6 and 9 (Stumm and Morgan, 1981, p. 121).

The **A**cid **N**eutralizing **C**apacity (ANC), often called alkalinity, determines the adsorption capability of acids in water and soils. It may be computed as follows:

$$ANC = C_b - C_a \quad (3.5)$$
$$C_b = K^+ + Na^+ + Mg^{2+} + Ca^{2+} + NH_4^+ + Al^{3+} \quad (3.6)$$
$$C_a = Cl^- + SO_4^{2-} + NO_3^- + F^- \quad (3.7)$$

where C_b is the sum of base cations resp. potassium, sodium, magnesium,

[9] note that 1 mol/l H^+ = 10^3 eq/m^3 H^+

calcium, ammonium, aluminum, and C_a is the sum of base anions resp. chloride, sulfate, nitrate and fluoride. In general the equations 3.6 and 3.7 may be extended with many more resp. H^+ -ion acceptors and H^+-ion donors.
Equations 3.1 to 3.7 describe the basics of acidification, further chemical elaboration of which will not be attempted in this study.

The comprehensive explanation by Eary et al. (1989) of the modeled processes in ETD, MAGIC and ILWAS has been summarized below in order to illustrate the line of thoughts in watershed acidification modeling. The summary presented here of Eary et al. (1989) is extended to indicate whether processes have been modeled in RAINS. The reader is referred to the original document (Eary et al., 1989) to obtain a full overview of details and of the technical, mostly chemically oriented, literature references.

The processes involved in watershed acidification (Figure 3.2) can broadly be classified in meteorological, hydrological, geochemical processes in soil and lake, and in biochemical processes.

Meteorology

Meteorologic parameters, e.g. precipitation and temperature, influence incoming water from rain and snowmelt and outgoing water from evapotranspiration. This is treated differently in the different models. ETD and ILWAS for example need daily meteorological data, MAGIC monthly or yearly averages and RAINS yearly value ranges.

The treatment of deposition chemistry is another important part of the modeled meteorology. Distinction is made between wet deposition (sulfate and other base ions in precipitation) and dry deposition (deposition on the surface due to diffusion and impaction in the absence of precipitation). MAGIC and ILWAS require data on a major part of the chemical elements in deposition. MAGIC uses a multiplication factor of wet deposition to compute dry deposition. ETD uses only ANC, $SO4^{2-}$ and Cl^-. RAINS inputs total (wet + dry) deposition, but computes the fraction of the sulfur flux to be actually used for the simulations.

Hydrology

Hydrological processes are concerned with the way in which precipitation is treated. The process is subdivided into surface hydrology, and subsurface hydrology, covering water flows on top of, and within the soils, and lake hydrology. ILWAS and MAGIC both take into account that a part of the precipitation is intercepted by the foliage of a forest canopy. RAINS distinguishes between deposition on open, i.e. agricultural areas including a lake, and forested regions. The assumption is made that forests intercept more pollutants than open areas. ETD, MAGIC and ILWAS take snowmelt into account when computing the net result of precipitation and evapotranspiration. The RAINS model does not explicitly simulate snowmelt and the pollutant storage capacity of snow.

In the subsurface hydrology all four models describe flow as a function of the hydraulic gradient and the hydraulic conductivity. For this purpose the soils are partitioned in different layers. RAINS distinguishes two such layers, i.e. the A-layer, consisting of the upper 50 cm of a soil, and the B-layer, for deeper soils.

In the lake hydrology the models differ in the assumptions with which incoming water is mixed with the entire lake volume.
MAGIC, ETD and RAINS assume perfect mixing. ILWAS allows for stratification of the lake into 80 layers. ILWAS, which is the by far most complicated model with respect to all modeled subprocesses, also takes heat exchange between the layers into account.

Geochemistry formulations

Geochemistry is concerned with the computation of ANC. Many chemical processes contribute to changes of ANC. In what follows a number of these processes are shortly clarified, since they are vital for an elementary understanding of soil and lake acidification.

1. ANC conventions: basically equation 3.5 is applied in all models although its form may differ and many more constituents may also be taken into account. An important issue in the computation of ANC is the way in which the Aluminum- water equilibrium is treated, which is basically described by

$$Al(OH)_3 + 3H^+ \longrightarrow Al^{3+} + 3H_2O \qquad (3.8)$$

The actual model formulation depends on the solubility constant, which is a function of temperature and the kind of gibbsite in the soil. The equilibrium stage is formalized as

$$[Al] = K_{s0} * [H^+]^3 \qquad (3.9)$$

where, K_{s0} is the solubility constant (mol/liter) with the following values:

$K_{s0} = 10^{8.11}$ for synthetic gibbsite,

$= 10^{8.77}$ for natural gibbsite, and

$= 10^{9.35}$ for microcrystalline gibbsite

Equation 3.9 may be extended on the right hand side with an aggregate of similar arguments to account for the different gibbsites.

2. Carbonic acid equilibria: the dissolution of carbon dioxide (CO_2) in water and the resulting carbonic acid specification, is what drives positive ANC (equation 3.5). All models treat this process as an equilibrium, as follows:

$$CO_2 + H_2O \longrightarrow HCO_3^- + H^+ \qquad (3.10)$$

The solubility of CO_2 depends on different constants i.e

$$K_1 = \{H^+\}*\{HCO_3^-\}/\{H_2CO_3\} \qquad (3.11)$$

$$K_h = \{H_2CO_3\}/pCO_2 \qquad (3.12)$$

where K_1 is called the first acidity constant and K_h is called the Henry's law constant. The buffering of H^+ ions by the exchange with base cations located on the soil surface, causes pCO_2, the partial CO_2 constant to increase. This results in an increase of the HCO_3^- concentration and of the ANC.

3. Organic acid chemistry: In this process the effects of generic organic acids on solution chemistry is considered. This process is taken into account by ILWAS in particular.

4. Mineral weathering: This process is concerned with the weathering reactions of silicate minerals in shallow soils and underlying exposed

bedrock. These minerals are formed under crustal conditions of high temperature and pressure and are subject to weathering reactions due to the thermodynamic instability of these silicates. The involved reactions consist of combinations of weak acids with strong bases. Weathering is highly dependent on the weathering rate:

$$R^w = k * (H^+) \qquad (3.13)$$

where,

R^w = weathering rate

k = rate constant for silicate hydrolysis

(H^+) = Hydrogen ion activity

Silicate hydrolysis can generally be formalized as

$$\text{Silicate Mineral} + H^+ \text{ --> Silica +/- Alumina + Cations} \qquad (3.14)$$

Although the basic equation used in ETD, MAGIC and ILWAS is similar to equation (3.13), the actual modeling of the weathering rate differs much among the models. One of the major reasons for that is the limited availability of field data, leading to MAGIC, ETD as well as ILWAS to apply a calibration procedure to match the weathering rate to particular field observations. Weathering rate in RAINS is one of the model parameters that is externalized by defining a range of values which is made subject to random sampling.

5. Cation exchange: This process involves the depletion of base cations by the consumption of H^+ ions. The process is much faster than the mineral weathering rate, resulting in the buffer capacity to become depleted at some point in time. The total capacity of base cations is called the Cation Exchange Capacity (CEC), and may consist of

$$CEC = Ca^{2+} + Mg^{2}_{+} + NH_4^+ + K^+ + Na^+ + H^+ \qquad (3.15)$$

The exchangeable fraction of base cations is called base saturation. MAGIC as well as ILWAS require estimates of CEC and base saturation that can be modified during model calibration, leading to accepted values that should lie within a reasonable range determined by field measurements. ETD calibrates the magnitude of CEC to result in a model response of ANC that matches ANC field measurements for the entire

watershed. RAINS has externalized CEC as well as base saturation.

6. Anion retention: SO_4^{2-} and NO_3^- anions may be adsorbed by the soils, a process that has been shown (see Eary et al., 1989) to be a function of pH. Sulfate concentration is a more important driver of acidification than Nitrate, due to the fact that nitrates tend to be taken up by vegetation. The concentration of Sulfate in the soil solution is however limited by the adsorption capacities of the soil minerals for SO_4^{2-}. The dependence of pH is modeled by neither of the models. Eary et al. (1989) argue that this lack may lead to an inaccurate description of Sulfate adsorption over long times during which significant changes in soil solution pH may occur. The pragmatic approach followed in MAGIC, ETD and ILWAS reduces the data requirements for sulfate adsorption capacity. Such data for a whole catchment, subcatchment or soil layers are hardly obtainable, since adsorption capacities and soil solution pH may vary aerially and with depth in a watershed. MAGIC uses a nonlinear relation between the equilibrium concentration of a sorbed anion in the soil, in the solution and the maximum sulfate adsorption capacity of soil. ETD and ILWAS both use linear relations to describe this process. The RAINS model has no representation of this process, simply because it is not observed in the northern part of Europe, for which the model has originally been designed.

Biogeochemical formulations

Biogeochemical processes involve the uptake of nutrients from soil layers by vegetation, decomposition of organic matter and the influence exerted on chemical reactions, by microbes. Three subprocesses are of importance :

1. Sulfate reduction: ANC in a lake may increase due to the reduction of SO_4^{2-}. The reduction process may be formalized as follows

$$SO_4^{2-} + 2CH_2O + 2H^+ \longrightarrow H_2S + 2CO_2 + H_2O \qquad (3.16)$$

Equation 3.16 shows that H^+ ions may be consumed due to sulfate reduction resulting in a net increase of ANC. All four models have modeled the process of sulfate reduction. MAGIC, ETD and ILWAS apply

differential equations that vary from one another by the use of rate constants and the inclusion of variables like lake depth and lake area. RAINS uses a linearized formula taking into account the catchment and lake area, using a stochastic parameter to simulate the fraction of the total SO_4^{2-} coming into a lake that is reduced. The external definition of such fraction is also part of the MAGIC simulation of the process.

2. Nitrification: this process describes the oxidation of ammonia which is mediated by microbes. ILWAS has the most extensive description of this process, followed by MAGIC that treats the loss of ammonia from the soil as a nutrient uptake process. ETD and RAINS have not incorporated this process.

3. Nutrient uptake: here the uptake of NO_3^-, NH_4^- and SO_4^{2-} by vegetation is represented. Both ILWAS and MAGIC have included this process, each in a different way as far as detail is concerned. RAINS and ETD do not simulate this process.

Other biogeochemical processes

The chemical processes addressed here are:

1. canopy-induced changes in throughfall (precipitation through the canopy) chemistry.
2. litter fall and decay, and
3. root respiration.

These detailed processes have only been represented in ILWAS and have not been included in ETD, MAGIC and RAINS.

The next section describes the equations that represent the above processes incorporated in the RAINS Lake Model.

3.3 THE RAINS LAKE MODULE EQUATIONS

Figure 3.3 displays the overall structure of the RAINS Lake Module. In this section the model equations representing the processes of section 3.2 in the RAINS Lake model are formalized.

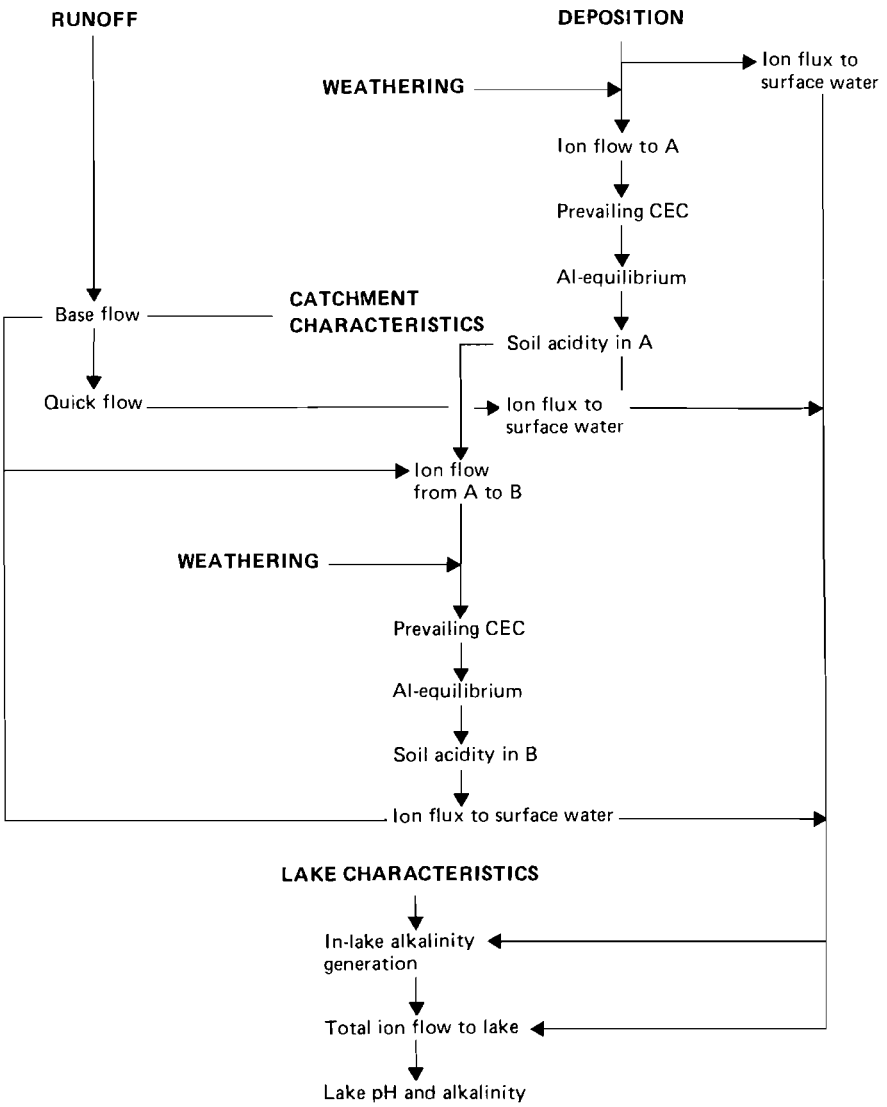

Figure 3.3 The structure of the RAINS Lake Module (source: Kämäri, 1988, p. 20).

The subscript t indicates the state of a model variable at simulation time t. The subscript 0 indicates the initial condition of a model variable.

Meteorology

Total deposition of sulfur (g/m^2_yr) is obtained from the EMEP long range transport model (Eliassen and Saltbones, 1983) and transformed to acid load (eq/m^2_yr).

First, the net acid load to the forest and open land is computed as the fraction of the total acid load that excludes base cations.

$$(D^{load})_t = (D^{tot})_t - (D^{bc})_t \qquad (3.17)$$

where,

$(D^{load})_t$ = the net acid load in year t in eq/m^2

$(D^{tot})_t$ = total deposition on the area in year t in eq/m^2

$(D^{bc})_t$ = the flux of base cations in year t in eq/m^2

It is assumed that more pollutants are filtered out by forests compared to open land:

$$(D^{loadf})_t = frac * (D^{loado})_t \qquad (3.18)$$

where,

$(D^{loadf})_t$ = acid load on forests in year t in eq/m^2

$(D^{loado})_t$ = acid load on open land in year t in eq/m^2

frac = forest filtering factor

The total deposition on the area is the sum of deposition on forests and on open land:

$$(D^{load})_t = f*(D^{loadf})_t + (1-f)*(D^{loado})_t \qquad (3.19)$$

where,

f = fraction of forests within the area

substitution of 3.17 in 3.18 gives:

$$(D^{loadf})_t = (D^{load})_t * frac/\ (1+(frac-1)*f) \qquad (3.20)$$

Hydrology

The hydrologic processes are dependent on monthly temperature and precipitation data as follows:

$$T(m) = T^{min}(m) + (T^{max}(m) - T^{min}(m)) * tfac \quad (3.21)$$

$$P(m) = P^{min}(m) + (P^{max}(m) - P^{min}(m)) * pfac \quad (3.22)$$

where,

$T(m)$ = the temperature in °C in month m

$T^{max}(m)$ = maximum temperature in °C in month m

$T^{min}(m)$ = minimum temperature in °C in month m

$P(m)$ = the precipitation in month m in meter (m)

$P^{max}(m)$ = maximum precipitation in month m in meter (m)

$P^{min}(m)$ = minimum precipitation in month m in meter (m)

m = month m, m = 1, ... ,12

pfac,tfac = random factors from the interval [0,1]

The evapotranspiration is computed from 3.21 as follows

$$E(m) = efac * T(m) \qquad \text{if } T(m) > 0 \quad (3.23)$$

where,

E(m) = evapotranspiration in month m (E(m) = 0 if $T(m) \leq 0$)

efac = an empirical constant

From 3.21, 3.22 and 3.23 the runoff is computed as

$$R = (P(1) - E(1)) + (P(2) - E(2)) + \ldots + (P(12) - E(12)) \quad (3.24)$$

where,

R = yearly runoff in m

In the subsurface hydrology runoff is used to compute the quickflow in the A-layer of the soil (top 0.5 m) and the baseflow in the B-layer (deeper than 0.5 m). The baseflow is computed according to catchment properties i.e. hydraulic conductivity, surface slope, catchment width and soil thickness of the B-layer:

$$B^{thick} = \max (S^{thick} - 0.5, 0) \quad (3.25)$$

where,

B^{thick} = soil thickness of the B-layer in m

S^{thick} = soil thickness in a region in m

The catchment properties are all sampled from an interval that is a reasonable representation of field measurements. The catchment area is computed as follows:

$$t^{catch} = a^{catch} - a^{lake} \quad (3.26)$$

where,

t^{catch} = terrestrial catchment area in m^2

a^{catch} = total catchment area in m^2

a^{lake} = lake area in m^2

Another catchment property is the catchment width. The width represents the circumference of the average terrestrial catchment, assuming a circular lake in a circular catchment.

$$C^{width} = (pi)^{1/2} * ((a^{catch})^{1/2} + (a^{lake})^{1/2}) \quad (3.27)$$

where,

C^{width} = width of the catchment in m

pi = 3.14159...

Baseflow is formulated from 3.24 to 3.27 as:

$$F^{b} = \min (C^{cond} * C^{slope} * C^{width} * B^{thick} / t^{catch} , R) \quad (3.28)$$

where,

F^{b} = baseflow in m

C^{cond} = hydraulic conductivity in the catchment in m/yr

C^{slope} = mean slope of the catchment in m/m

From 3.28 and 3.24 the quickflow is computed as:

$$F^{q} = R - F^{b} \quad (3.29)$$

where,

F^{q} = quickflow in m

The total water volume entering the lake becomes:

$$L^{vol} = R * a^{catch} \quad (3.30)$$

where,

Lvol = water volume entering the lake in m^3

The water volume contained in the lake is expressed as

$$L^{mix} = L^{depth} * a^{lake} \quad (3.31)$$

where

L^{mix} = volume of the lake in m^3

L^{depth} = mean lake depth in m

Geochemical processes

The following set of equations represent the geochemical processes. These processes take place in the A- as well as in the B-layer. First buffer capacity and silicate buffer rate are computed as function of the thickness of the A-layer:

$$A^{thick} = \min (S^{thick}, 0.5) \quad (3.32)$$

where,

A^{thick} = soil thickness of A-Layer

The calcium buffer capacities in A- and B-layer thus become

$$(A^{cap})_0 = a^{cap} * A^{thick} \quad (3.33)$$

$$(B^{cap})_0 = b^{cap} * B^{thick} \quad (3.34)$$

where,

$(A^{cap})_0$ = Calcium buffer capacity in eq/m^2 (A-layer) in year t=0

a^{cap} = sampled Calcium buffer capacity in eq/m^3 (A-layer)

$(B^{cap})_0$ = Calcium buffer capacity in eq/m^2 (B-layer) in year t=0

b^{cap} = sampled Calcium buffer capacity in eq/m^3 (B-layer)

The silicate buffer rates in both layers become:

$$A^{rate} = S^{rate} * A^{thick} \quad (3.35)$$

$$B^{rate} = S^{rate} * B^{thick} \quad (3.36)$$

where,

A^{rate} = silicate buffer rate in eq/m^2_yr (A-layer)

S^{rate} = sampled Silicate buffer rate in eq/m^3_yr (in the soil)

B^{rate} = silicate buffer rate in eq/m^2_yr (B-layer)

The cation exchange capacity expressions used are:

$$A^{cec} = S^{cec} * A^{thick} \quad (3.37)$$

$$B^{cec} = S^{cec} * B^{thick} \quad (3.38)$$

where,

A^{cec} = cation exchange capacity in eq/m^2 (A-layer)

B^{cec} = cation exchange capacity in eq/m^2 (B-layer)

S^{cec} = sampled cation exchange capacity in eq/m^3 in the soil

A fraction of the cation exchange capacity is the base saturation:

$$(A^{sat})_0 = a^{sat} * A^{cec} \quad (3.39)$$

$$(B^{sat})_0 = b^{sat} * B^{cec} \quad (3.40)$$

where

$(A^{sat})_0$ = actual CEC in eq/m^2 (A-layer) in year t=0

a^{sat} = sampled base saturation (A-layer) when t=0.
If t > 0 then $a^{sat} = (A^{sat})_t \ / \ A^{cec}$

$(B^{sat})_0$ = actual CEC in eq/m^2 (B-layer) in year t=0

b^{sat} = sampled base saturation (B-layer) when t=0.
If t > 0 then $b^{sat} = (B^{sat})_t \ / \ B^{cec}$

Finally the field capacity, the maximum amount of water that the unsaturated zone of a soil can hold against the pull of gravity, in the A and B layer is computed as:

$$f^{capA} = f^{cap} * A^{thick} \quad (3.41)$$

$$f^{capB} = f^{cap} * B^{thick} \quad (3.42)$$

where,

f^{capA} = field capacity in m (A-layer)

f^{capB} = field capacity in m (B-layer)

f^{cap} = sampled field capacity fraction

The following equations describe the chemical reactions that lead to a change of $[H^+]$ and the alkalinity. First the initial condition of lake alkalinity and soil pH is defined:

$$(L^{alk})_0 = S^{thick} * S^{rate} * t^{catch} / (R * a^{catch}) \quad (3.43)$$

$$(H^{+A})_0 = c \quad (3.44)$$

$$(H^{+B})_0 = c \quad (3.45)$$

$$(L^{H+})_0 = K_1 * K_H * pCO_2 / (L^{alk})_0 \quad (3.46)$$

where,

$(L^{alk})_0$ = initial lake alkalinity in year t=0 in eq/m^3

$(H^{+A})_0$ = initial Hydrogen ion concentration in year t=0 (A-layer) in eq/m^3

$(H^{+B})_0$ = initial Hydrogen ion concentration in year t=0 (B-layer) in eq/m^3

c = starting constant (10^{-4} eq/m^3 which equals pH=7)

$(L^{H+})_0$ = initial Hydrogen concentration in the lake in year t=0 in eq/m^3

K_1 = first acidity constant in eq/m^3

K_H = Henry's law constant in eq/m^3_atm

pCO_2 = partial CO_2 pressure in atm

Now the iterative representation of chemical reactions in soil layers and in the lake can start.

Soil chemistry

In each soil layer a three step buffering sequence is simulated by resp. the:

1. Calcium carbonate buffer range
2. Silicate and Cation Exchange range
3. Aluminum range

First the buffer sequence in the A-layer is simulated starting with the carbonate range. Equations 3.47a and 3.47b are only simulated if condition 3.47 is met:

$$(A^{cap})_{t-1} \geq 0 \quad (3.47)$$

$$(A^{cap})_t = \max\ ((A^{cap})_{t-1} - (D^{loadf})_t\ ,\ 0) \qquad (3.47a)$$

$$(H^{+A})_t = c_1 \qquad (0.000631 \text{ which equals a pH = 6.2}) \qquad (3.47b)$$

If 3.47 is not met, the net acidifying potential of the A-layer is computed as:

$$(A^{load})_t = (D^{loadf})_t - (A^{rate})_t \qquad (3.48)$$

where,

$(A^{load})_t$ = acidifying potential in eq/m^2_yr

and the next buffering range is entered, i.e the silicate and cation exchange buffer range, only if condition 3.49 is met:

$$(H^{+A})_{t-1} < 0.1\ eq/m^3\_yr \qquad (\text{equivalent to } pH_{t-1} > 4) \qquad (3.49)$$

Then, if there is cation exchange capacity left in the A-layer, it is exhausted with a rate equal to the potential defined in 3.47, as follows

$$(A^{sat})_t = \max\ (\ \min\ ((A^{sat})_{t-1} - (A^{load})_t,\ A^{cec}),\ 0) \qquad (3.49a)$$

A functional relationship between the base saturation and the pH leads to the computation of the hydrogen ion concentration (see Reuss, 1980):

$$(H^{+A})_t = 10\ **\ (-1 - 1.6*\ ((A^{sat})_t\ /\ A^{cec})^{0.75}) \qquad (3.49b)$$

where ** denotes the power operator.

If 3.49 is not met, thus the $pH_{t-1} \leq 4$ and there is no cation exchange capacity left in the soil, the computation of Hydrogen ion concentration results from an <u>equilibrium</u> relationship between $[H^+]$ and the aluminum ion concentration, $[Al^{3+}]$, assuming a certain solubility of a solid phase of aluminum:

$$(Al^{3+A})_{t-1} = K_{s0} * ((H^{+A})_{t-1})^3 \qquad (3.50)$$

where,

$(Al^{3+A})_{t-1}$ = Aluminum equilibrium concentration in year t-1 in the A-layer in eq/m^3

K_{s0} = gibbsite solubility constant

The dissolution or precipitation of aluminum takes place until the gibbsite equilibrium stage is reached. Precipitation that infiltrates the soil leads to a change of the disequilibrium concentrations of aluminum and hydrogen (see Kämäri, 1988, pp. 25-26):

$$(H^{+Ad})_t = (F^{capA} * (H^{+A})_{t-1} + (A^{load})_t)/(F^{capA} + R) \quad (3.51)$$

$$(Al^{3+Ad})_t = (F^{capA} * (Al^{3+A})_{t-1})/(F^{capA} + R) \quad (3.52)$$

where,

$(H^{+Ad})_t$ = disequilibrium hydrogen ion concentration in year in eq/m^3

$(Al^{3+Ad})_t$ = disequilibrium aluminum ion concentration in year t in eq/m^3

The disequilibrium relation is formalized as follows:

$$(Al^{3+Ad})_t - (Al^{3+A})_t = (H^{+A})_t - (H^{+Ad})_t \quad (3.53)$$

Substitution of 3.50, 3.51 and 3.52 in 3.53, leads to a cubic equation which has a single real root (Abramowitz and Stegun, 1965, p.17, 3.8.2) of the hydrogen ion concentration , that will be expressed below using four auxiliary variables h_1, h_2, h_3 and h_4:

$$h_1 = (H^{+Ad})_t + (Al^{3+Ad})_t \quad (3.54)$$

$$h_2 = 1 / (3 * K_{s0}) \quad (3.55)$$

$$h_3 = -h_1 / (2 * K_{s0}) \quad (3.56)$$

$$h_4 = ((h_3)^2 + (h_2)^3)^{1/2} \quad (3.57)$$

$$(H^{+A})_t = (-h_3 + h_4)^{1/3} - (h_3 + h_4)^{1/3} \quad (3.58)$$

The soil is assumed to be controlled by the aluminum solubility as long as the weathering rate is not able to produce excess base cations i.e $(A^{load})_t \leq 0$. If however condition 3.49 is fulfilled and $(A^{load})_t > 0$, then $(H^{+A})_t$ has been set to 0.1 which is equivalent to a soil pH of 4. Finally the flux of H^+ ions that is released from the A-layer is computed:

$$(H^{fluxA})_t = F^q * (H^{+A})_t \quad (3.59)$$

where,

$(H^{fluxA})_t$ = flux of H^+ ions in eq/m^2_yr (A-layer)

The flux of bicarbonate ions, $(HCO_3)^-$, released from the A-layer becomes:

$$(C^{fluxA})_t = \max\ (A^{rate} - (D^{loadf})_t, 0) \qquad (3.60)$$

where,

$(C^{fluxA})_t$ = flux of bicarbonate ions in eq/m^2_yr (A-layer)

Now that all processes in the A-layer have been simulated, the next set of simulations relate to the B layer. The process equations are basically a repetition of equations 3.47 until 3.58 in which the A's should be replaced by B's. The transfer of the hydrogen ion concentration to the B-layer is simulated by:

$$(B^{stress})_t = (H^{+A})_t * F^b \qquad (3.61)$$

where,

$(B^{stress})_t$ = the net acid load coming from the A-layer in eq/m^2_yr (B-layer)

This influences the simulation of the carbonate buffer range in the B-layer (see equation 3.47a):

$$(B^{cap})_t = \max\ ((B^{cap})_{t-1} - (B^{stress})_t\ ,\ 0) \qquad (3.47aB)$$

Similarly 3.48 changes to:

$$(B^{load})_t = (B^{stress})_t - (A^{rate})_t \qquad (3.48B)$$

Other equations used in the A-layer that are to be changed more than just typographically, are related to the usage in the B-layer of the baseflow rather than the runoff. This influences the type of equation to replace 3.51 and 3.52 as follows:

$$(H^{+Bd})_t = (F^{capB} * (H^{+B})_{t-1} + (B^{load})_t)/(F^{capB} + F^b) \qquad (3.51B)$$

$$(Al^{3+Bd})_t = (F^{capB} * (Al^{3+B})_{t-1})/(F^{capB} + F^b) \qquad (3.52B)$$

Finally the hydrogen ion and bicarbonate ion concentration that are released by the B-layer are represented as:

$$(H^{fluxB})_t = F^b * (H^{+B})_t \quad (3.62)$$

$$(C^{fluxB})_t = \max\,(B^{rate} - (B^{stress})_t\ ,0) \quad (3.63)$$

where,

$(H^{fluxB})_t$ = flux of H^+ ions in year t in eq/m^2 (B-layer)

$(C^{fluxB})_t$ = flux of bicarbonate ions in year t in eq/m^2 (B-layer)

Lake chemistry

The last step consists of the simulation of the lake acidity. The in-lake alkalinity generation (Baker et al., 1986) from SO_4 retention in lakes is obtained from the total sulfur load:

$$(C^{fluxL})_t = L^{rate} * (D^{tot})_t \ / \ ((L^{depth} * R * a^{catch} / L^{mix}) + L^{rate}) \quad (3.64)$$

where,

$(C^{fluxL})_t$ = bicarbonate concentration released in the lake in year t in eq/m2

L^{rate} = sampled in-lake retention coefficient in m/yr

The total hydrogen ion load in the lake can now be computed from 3.59, 3.62 and the direct input from precipitation on the lake:

$$(H^{fluxT})_t = ((H^{fluxB})_t + (H^{fluxA})_t) * t^{catch} + (D^{loado})_t * a^{lake} \quad (3.66)$$

where,

$(H^{fluxT})_t$ = total hydrogen ion load in the lake in year t in eq

The total bicarbonate ion load in the lake is similarly computed from 3.60 and 3.63 and the internal alkalinity generation in the lake:

$$(C^{fluxT})_t = ((C^{fluxA})_t + (C^{fluxB})_t) * t^{catch} + (C^{fluxL})_t * a^{lake} \quad (3.67)$$

where,

$(C^{fluxT})_t$ = total bicarbonate ion load in the lake in year t in eq

The equilibrium relationship between $[H^+]$ and $[HCO_3^-]$ is as follows (see also equations 3.10,3.11 and 3.12)

$$(L^{alk})^t = K_1 * K_H * pCO_2 / (L^{H+})_t \quad (3.68)$$

where,

K_1 = first acidity constant in eq/m^3

K_H = Henry's law constant in eq/m^3_atm

pCO_2 = partial CO_2 pressure in atm.

Because of the inflow of hydrogen and bicarbonate ions, disequilibrium relations persevere:

$$(H^{fluxTd})_t = ((H^{fluxT})_t + (L^{H+})_{t-1} * L^{mix}) / (L^{mix} + L^{vol}) \quad (3.69)$$

$$(C^{fluxTd})_t = ((C^{fluxT})_t + (L^{alk})_{t-1} * L^{mix}) / (L^{mix} + L^{vol}) \quad (3.70)$$

where,

$(H^{fluxTd})_t$ = disequilibrium load of Hydrogen ion in year t in eq/m^3

$(C^{fluxTd})_t$ = disequilibrium load of bicarbonate ion in year t in eq/m^3

$(L^{H+})_{t-1}$ = Hydrogen ion concentration in the lake in year t-1 in eq/m^3

$(L^{alk})_{t-1}$ = bicarbonate ion concentration (alkalinity) in the lake in year t-1 in eq/m^3

The disequilibrium relation, which represents that equal amounts of hydrogen and bicarbonate ions are consumed, is formalized as follows:

$$(L^{H+})_t - (H^{fluxTd})_t = (L^{alk})_t - (C^{fluxTd})_t \quad (3.71)$$

Substitution of 3.68, 3.69 and 3.70 in 3.71 leads to a second order equation for which the positive root for the bicarbonate concentration is accepted. The algebraic result, after introducing auxiliary variable h_5, h_6, h_7 and h_8 becomes:

$$h_5 = ((C^{fluxTd})_t - (H^{fluxTd})_t \quad (3.72)$$

$$h_6 = L^{mix} * ((L^{alk})_{t-1} - (L^{H+})_{t-1}) \quad (3.73)$$

$$h_7 = L^{mix} + L^{vol} \quad (3.74)$$

$$h^8 = 0.5 * (h_5 + h_6) / h_7 \quad (3.75)$$

$$(L^{alk})_t = h_8 + ((h_8)^2 + K_1 * K_H * pCO_2)^{1/2} \quad (3.76)$$

$$(L^{H+})_t = K_1 * K_H * pCO_2 / (L^{alk})_t \quad (3.77)$$

Equation 3.76 is the lake-alkalinity in year t and 3.77 represents the Hydrogen ion concentration of the lake in year t. The pH is computed by substitution of 3.77 in equation 3.4. The pH will be the model response variable that is analyzed in the remainder of the study.

3.4 SPATIAL GENERALIZATION

The MAGIC, ILWAS , ETD and RAINS Lake models have all been verified against field measurements of particular catchments. Regionalization, however, consists of applying the models to an entire region[10] in contrast to a few individual lakes. MAGIC has been calibrated to a region by means of the regional sensitivity method (HSY algorithm) that has been introduced in chapter 2, using Monte Carlo.

The regional version of ILWAS (RILWAS, see Goldstein et al., 1984) consists of defining typical parameter values that are assumed to be representative of lakes in a region.

By using the ETD model in conjunction with multiple regression on lake data about watershed variables (exogenous) and alkalinity (endogenous), regional forecasts of lake resources at risk were obtained (Schnoor et al., 1986). Regionalization of the RAINS Lake Module has been achieved in the RAINS model by predefining 14 lake regions in northern Europe (Scandinavia). For each of the regions, model parameter value ranges have been defined to which a filtering procedure (see chapter 2) is applied in combination with Monte Carlo simulation.

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[10]Note that in the economic sciences the term regionalization often refers to subdividing a larger area into smaller ones (top down). In this study, regionalization consists of defining larger areas from single watersheds (bottom up).

3.5 CONCLUDING REMARKS

Many interrelated meteorological, hydrological, soil surface, soil chemical and lake chemical processes (Figure 3.2) have been identified to contribute to watershed acidification. Modeling of watershed acidification has therefore not only been concerned with the definition of the relative importance of each of these processes but also with its formal representation. Many different models have been built. A short description of the modeled processes has been presented in conjunction with three currently well known models i.e the ILWAS, the ETD and the MAGIC model.

A detailed mathematical description of the difference equations representing the processes that are simulated in the RAINS model was provided.

The major problem of the modeling efforts has been the lack of data, especially when results have to be obtained in order to describe acidification processes in regions containing lakes. Calibration procedures have therefore taken an important place in watershed acidification modeling. Calibration results are however dependent on the choice of regional boundaries, as will be shown in the following chapter. Thus, one of the products of this study is a concept, based on uncertainty analysis, in which both the calibration effects and the model application to predefined regions are relaxed.

In the next chapter a calibration procedure is introduced that, instead of using all model parameters, only uses the ones that are shown to have the largest influence on the variability of model predictions. The issue in chapter 4 is addressed whether a calibration method for model applications in regions, of which the boundaries have been predefined, affects the confidence of model predictions.

4. REGIONAL VALIDATION OF THE RAINS LAKE MODEL

4.1 INTRODUCTION

The definition of a region (see chapter 1) and its boundaries is often based on historical, pragmatic or political precedents. Such established regional boundaries often tend to serve as a convenient spatial scale within which issues can be investigated, but for which the region was not otherwise distinguished. An example is the application of the REgional Soil Acidification Model (RESAM; see de Vries, 1987) to COROP regions in the Netherlands. The Netherlands distinguishes 40 COROP regions (CBS, 1975) based on a categorization of social-economic functional relationships, not taking any environmental consideration into account. The policy assessment of the state of large scale environmental subsystems, e.g. water quality, effects of deposition and of climatic change is dependent on the scale at which data has been collected and aggregated. The analysis of large scale systems often involves the application of a model to a study area that has been partitioned into zones. The choice of the zoning system often leads to the aggregation of data to match the chosen scale. There are many ways in which zones can be chosen and aggregation levels defined. It has been shown in the field of spatial interaction modeling, e.g. models in which inter zonal flows are described that the modeling results are not invariant to the choice of the zonal boundaries (Openshaw, 1977a; 1977b; 1978; Openshaw and Taylor, 1981).

The objective of this chapter is to determine the effect that predefined regional boundaries have on the estimate of broad scale environmental effects using a model that is a function of site specific estimates. More specifically, it will be shown that the calibration of the model output to regional measurements, may affect the confidence of model predictions. The investigation concentrates on the RAINS Lake Module. In order to estimate the influence of model complexity also a metamodel of the RAINS Lake Model is applied. The spatial scale of both model applications is chosen similar to the scale of the RAINS Lake

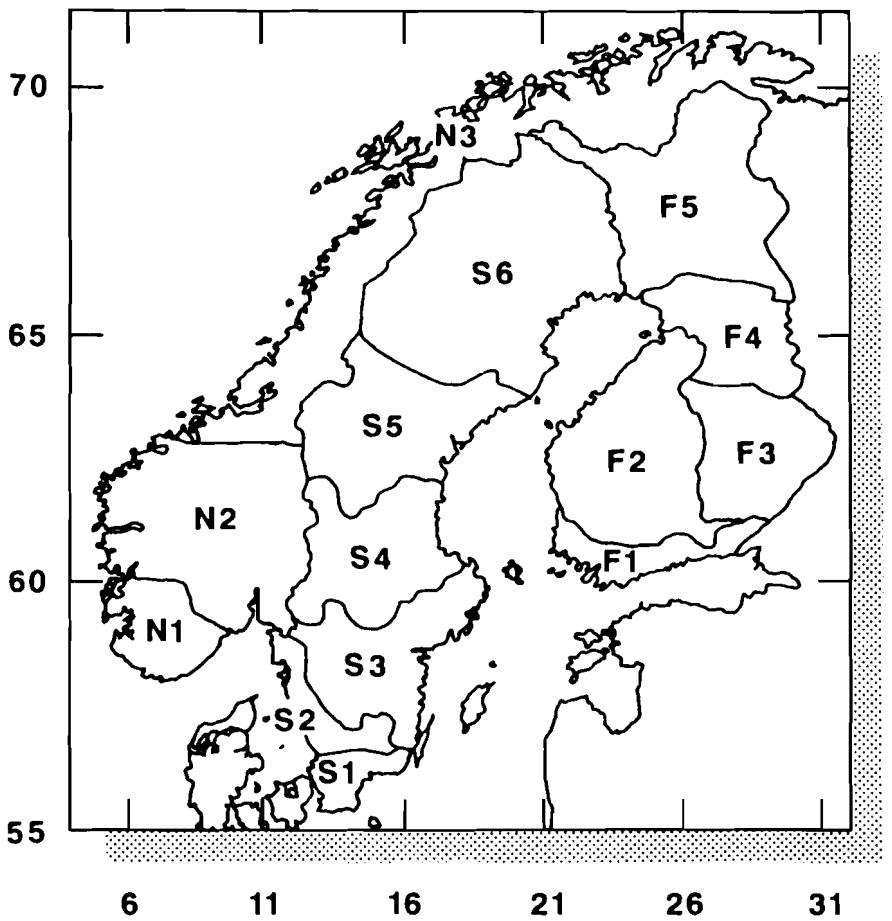

Figure 4.1 14 lake regions demarcated for the usage of the RAINS Lake Module.

Model that was distinguished in the RAINS model. This scale was restricted to Northern Europe, i.e. Scandinavia. The reason for restricting the RAINS Lake Module to that area was (1) the large amount of lakes in that area (2) the early policy involvement leading to the support of monitoring programs and (3) the available data resulting from these monitoring programs (see section 4.2). A total of 14 lake regions was chosen in Scandinavia (see Figure 4.1). The regions in Finland were distinguished on the basis of criteria about soil sensitivity (Kämäri, 1988, pp 8-15). Sensitive areas are ranked as result of the selection of terrestrial and aquatic sites where atmospheric deposition may have the largest unfavorable effects. The prediction of the extent and severity of the effects is not the purpose of ranking sensitive areas. Policy makers are predominately interested in forecasting environmental effects. However, the policy evaluation of the effect of emission abatement strategies in regional environmental systems, e.g. watershed acidification, may suffer from uncertainties that are introduced by the combination of a zonal system and a model. This is shown in the following.

First the way in which the RAINS Lake Module is driven and calibrated is discussed.

4.2 The RAINS LAKE MODULE; DRIVER, DATA AND CALIBRATION

As alluded to in chapters 2 and 3, the RAINS Lake Module is driven in a Monte Carlo simulation framework. Two such shells have been built around the module. The first one (see the filtering procedure in chapter 2) does not incorporate model parameter uncertainties, whereas the second, used in this study, does. Taking the parameter uncertainties into account is of importance especially with respect to calibration, as will be explained later.

The driver

The driver used in this study consists of a three stage procedure:

1. Monte Carlo sampling

2. running the model
3. model response analysis

The method of treating these three stages independently with respect to computer implementation, is available as a software package called PRISM (Gardner, 1983). The three stages are respectively called PRISM1, PRISM2 and PRISM3 which are linked to one another by file handling routines (Figure 4.2). In Figure 4.2 the RAINS Lake Module has been substituted in PRISM2, which is a model driver in which any model can be incorporated. PRISM1 reads the file (data.dat) of parameter value ranges. It performs a Latin Hypercube sampling procedure (chapter 2) which optionally allows for the implementation of rank correlations among the parameters (see Iman and Conover, 1982). The samples are written in a file (prism1.dat) which becomes input to the model in PRISM2.

PRISM2 produces an output file (prism2.dat) which can be used for the purpose of model response analysis. The latter may consist of different routines, the set of which is called PRISM3.

The decision of which parameters to include in the sampling file (data.dat) depends on the available data and on the variables (Monte Carlo parameters) that drive the model.

Table 4.1 lists the model variables that have been used as Monte Carlo parameters in data.dat of several model analyses (Kämäri et al., 1986, Gardner et al., 1987, Hettelingh and Gardner, 1988, Hettelingh et al., 1988).

The number of Monte Carlo simulations proved sufficient around 500 (Gardner et al., 1980a; O'Neill et al., 1982). The criteria for testing the appropriateness of the number of runs was to apply the Kolmogorov-Smirnov test on the cumulative distributions of model responses, i.e. pH, of two subsequent runs. No significant difference at a 5% confidence level usually ensued before 500 runs. Another criteria consisted of verifying whether the correlations between the sampled input parameter values, were small (< 0.001). This criteria was equally met at a number of runs smaller than 500.

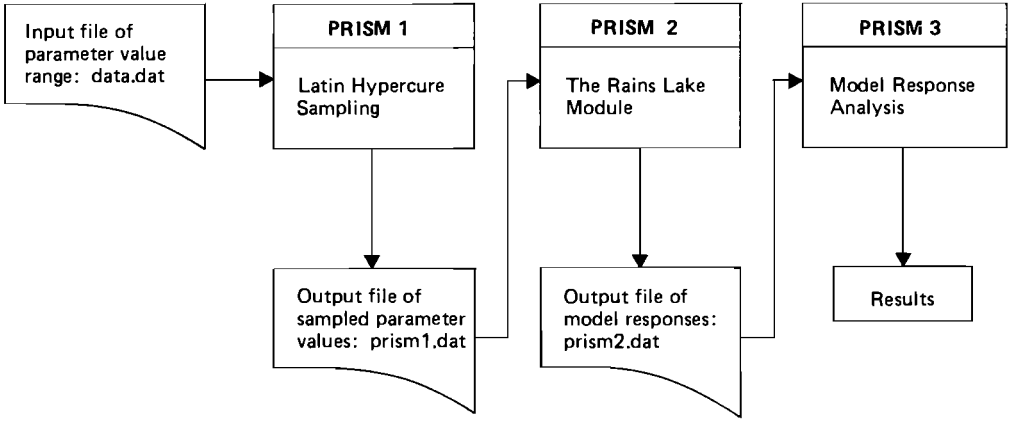

Figure 4.2 The PRISM shell for the performance of sensitivity and uncertainty analysis of a model.

Table 4.1: Monte Carlo parameters for the RAINS Lake Module[1]

Symbol (chapter 3) (equation #) (first used)	Code name[2]	Short description
a^{lake} (3.26)	lakar	lake area
a^{catch}/a^{lake} (3.26)	clrat	catchment to lake ratio
L^{depth} (3.31)	ldept	lake depth
S^{thick} (3.25)	soilt	average soil thickness
C^{slope} (3.28)	slope	slope of the catchment
C^{cond} (3.28)	cond3	hydraulic conductivity
S^{rate} (3.35)	sibr	silicate buffer rate
S^{cec} (3.37)	cec	cation exchange capacity
a^{sat} (3.39)	bsata	base saturation in A-layer
f^{cap} (3.41)	fcap	soil moisture content
b^{sat} (3.40)	bsatb	base saturation in B-layer
$L^{alk}*L^{H+}$ (3.68)	calk[3]	alkalinity constants
efac (3.23)	evs[3]	evapotranspiration const.
f (3.19)	satu	forest coverage
L^{rate} (3.64)	xso4	sulfur retention coeff.
frac (3.18)	fofi	forest filtering factor
D^{load}/D^{tot} (3.17)	1-sigma	base cation fraction
tfac (3.21)	tfac[4]	temperature factor
pfac (3.22)	pfac[4]	precipitation factor
D^{tot} (3.17)	sy	total deposition

[1]The number of input files of Scandinavia is equal to the number of regions, i.e. 14. A short description of the structure of an inputfile is given in appendix II.
[2]The code names refer to the names of the computer program variables. A description of the computer program can be found in appendix II.
[3]In some of the investigations a fixed cond-, calk- and evs-parameter was used.
[4]In some of the investigations a monthly temperature and precipitation range was added in data.dat (see equation 3.24), referred to as T(1), ... ,T(12) and P(1), ... ,P(12).

Sulfur deposition scenario

The meaning of the parameters of Table 4.1 in the context of the RAINS Lake Model, is extensively delineated in the equations of chapter 3. The treatment of deposition as random parameter needs some further explanation, since RAINS computes the deposition in every grid[11]. With respect to a lake region however, site specific levels of deposition are computed as random stress, say sy_0 which lies between the minimum

[11] The european area considered is bound by -12° longitude, 35° latitude and 42° longitude, 74° latitude. The grid size is 1° longitude by 0.5° latitude.

and maximum deposition levels in the region.

Note that simulations do not address particular lakes in particular grids, but are directed towards watershed site characteristics, in a region. By sampling from ranges of the parameters representing the watershed characteristics (Table 4.1), a representation is obtained of the kind of lakes 'typical' for that region. Monte Carlo Simulations thus result in 500 'typical' lakes.

The analysis in this chapter first treats the RAINS time horizon from 1960 to 2040, in which years regional deposition is assumed to follow the same path as the minimum and maximum deposition, starting from sy_0 in 1960. The deposition paths result from two RAINS scenarios i.e a high-emission-scenario and a low-emission-scenario, that deviate from one another from 1980 to 2040. The investigated deposition pattern resulting from both emission scenarios were computed with the EMEP-I source receptor matrix (Eliassen and Saltbones, 1983) and are resp. displayed in Figure 4.3 and Figure 4.4. Deposition becomes as high as 3 $g/m^2/yr$ (Figure 4.3) in the southern part of Sweden due to the high-emission-scenario. The low-emission scenario leads to 1 $g/m^2/yr$ (Figure 4.4) being the highest deposition level in Scandinavia. The deposition pattern in 1980 is addressed further below (Figure 4.7).

A second approach (section 4.4), applied to a metamodel, consists of considering European emission history (Fjeld, 1976, cited in Whelpdale, 1987) starting from 1920, and allowing yearly deposition in future years to be randomly drawn between the minimum and maximum deposition in a region.

The data

Field measurements of watershed characteristics and pH and alkalinity levels were sampled in 5 regions of Finland, 6 regions of Sweden and 3 regions of Norway. For each lake region a set of corresponding parameter distributions was developed based on these measurements. The quality and quantity of the data on which these input distributions are based varied for all regions of the three countries. Table 4.2 illustrates the differences in sampling intensities between the regions.

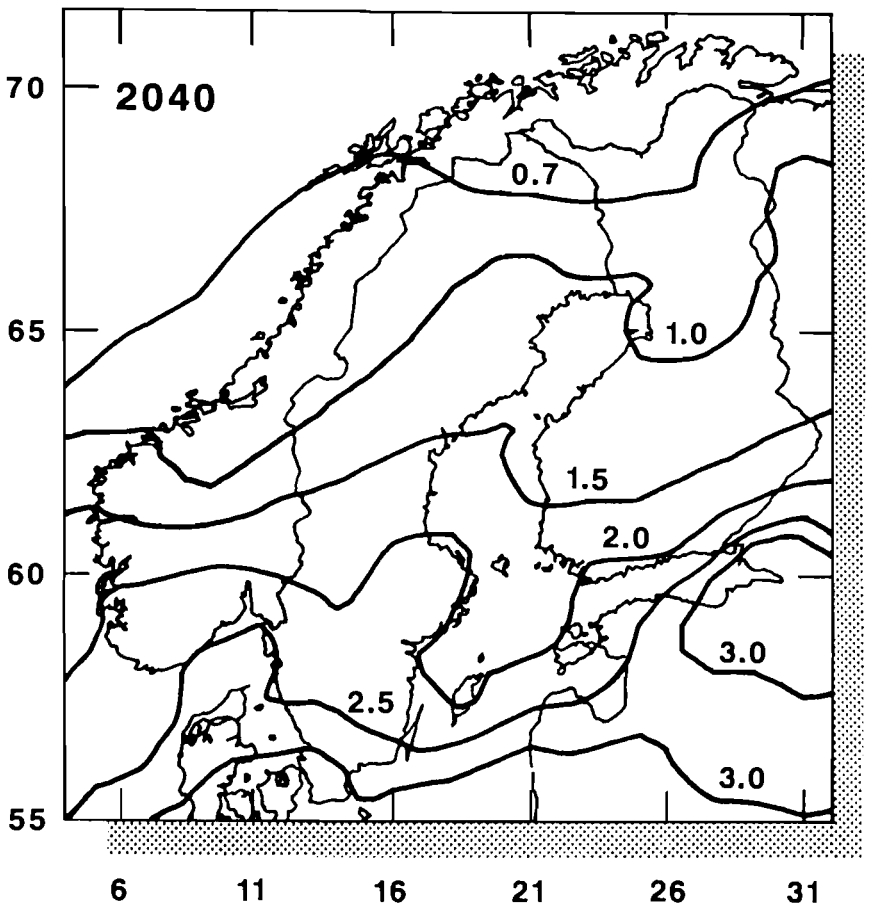

Figure 4.3 The deposition pattern over Scandinavia in 2040 as result of the High Deposition Scenario

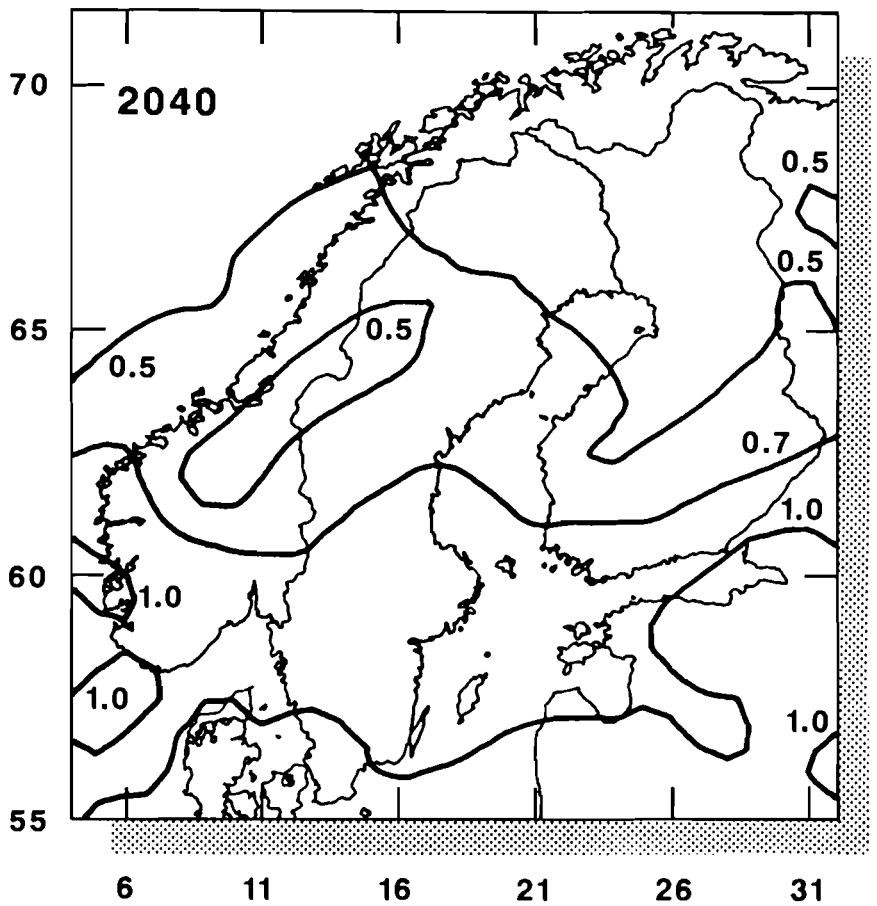

Figure 4.4 The deposition pattern over Scandinavia in 2040 as result of the Low Deposition Scenario

Table 4.2 Watersheds sampled in 14 lake regions[1]

Country Region	Total Number of Lakes	Number[3] of Lakes Sampled	Percent of Lakes Sampled
Finland			
1	2833	1590	56.1
2	13579	647	4.8
3	12146	2061	16.9
4	9644	1537	15.5
5	17841	512	2.9
Norway			
1	}	382	}
2	} unknown[2]	65	} unknown
3	}	67	}
Sweden			
1	1194	}	15% lakes 1-9 ha
		}	50% lakes 10-99 ha
2	5887	}	}
3	7580	} 8000	}
4	15308	}	} 2% of all
		}	} Swedish lakes
5	9943	}	} have been
		}	} sampled
6	47500	}	}

[1] See Fig. 4.1 for geographical location of regions in Scandinavia.
[2] The total number of lakes in Norway is approximately 3000.
[3] The Finnish lakes were sampled under supervision of the Finnish National Board of Waters and Environment between 1975 and 1983. The Swedish morphological data was obtained from the Swedish Meteorological and Hydrological Institute (SMHI) and the information on the lake acidification was obtained from the National Swedish Environmental Protection Board from samples taken between 1977 and 1980. The data used here was obtained from reports which aggregated the information into frequency distributions in which details about the sample sizes were made explicit. The Norwegian data were sampled by the Norwegian Institute for Water Research (NIVA) between 1974 and 1977.

Uncertainty analysis and calibration

No calibration would be necessary if the model were known to correctly represent the behavior of each catchment and if the a priori information on the shape of the parameter distributions, that were derived from the measurements, were correct. The model would then produce reliable pH frequency distributions. The data, however, as illustrated by Table 4.2, is characterized by a high degree of heterogeneity and generalization. The result of not applying a

calibration method is shown in Figure 4.5.

Figure 4.5 displays the cumulative frequency distribution of pH model response in 1920 and 1980, using the European sulfur emission history as mentioned above, compared to the cumulative frequency distribution of measured pH in 1980 in region 1 of Finland. The cumulative distribution of 1920 has been displayed to show the models initial response as compared to the last simulation year 1980. As can be seen, only about 5% of the measurements have a pH lower than 5, whereas Monte Carlo simulation leads to about 20% of the lakes with a pH less than 5 in 1980. Evidently the lower pH ranges are being overestimated by the model without calibration.

A model calibration procedure was designed for this study that (1) was simple to apply, (2) resulted in adequate model prediction of the measured frequency distribution of 1980 watershed pH, and (3) produced parameter sets that could be used in other regions, and (4) allows for the estimation of uncertainties of model applications as function of predefined regions.

The calibration procedure was applied to the southern parts of the three countries in Scandinavia because these regions receive the highest levels of sulfur deposition. The calibration method consists of changing the shapes of the frequency distributions assigned to parameters most relevant[12] for the explanation of the pH model response variation. A flowchart of the calibration procedure is provided in Figure 4.6.

An uncertainty analysis investigating partial R^2 (see equation 2.16) leads to a parameter ranking in which the soil thickness (soilt) and silicate buffer rate (sibr) were most important. The results of the uncertainty analysis are displayed in Table 4.3.

The definition of calibration given in section 2.4 involves a recursive modification of model parameters until the comparison of model response and measurements agrees with a goodness of fit criterion. Traditional tests of goodness-of-fit, such as the Kolmogorov-Smirnov or Chi-squared test, are not well suited to this purpose because significant differences may result that are irrelevant with respect to acidity.

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[12]A parameter is more relevant than another if its partial R^2 is higher (see section 2.4).

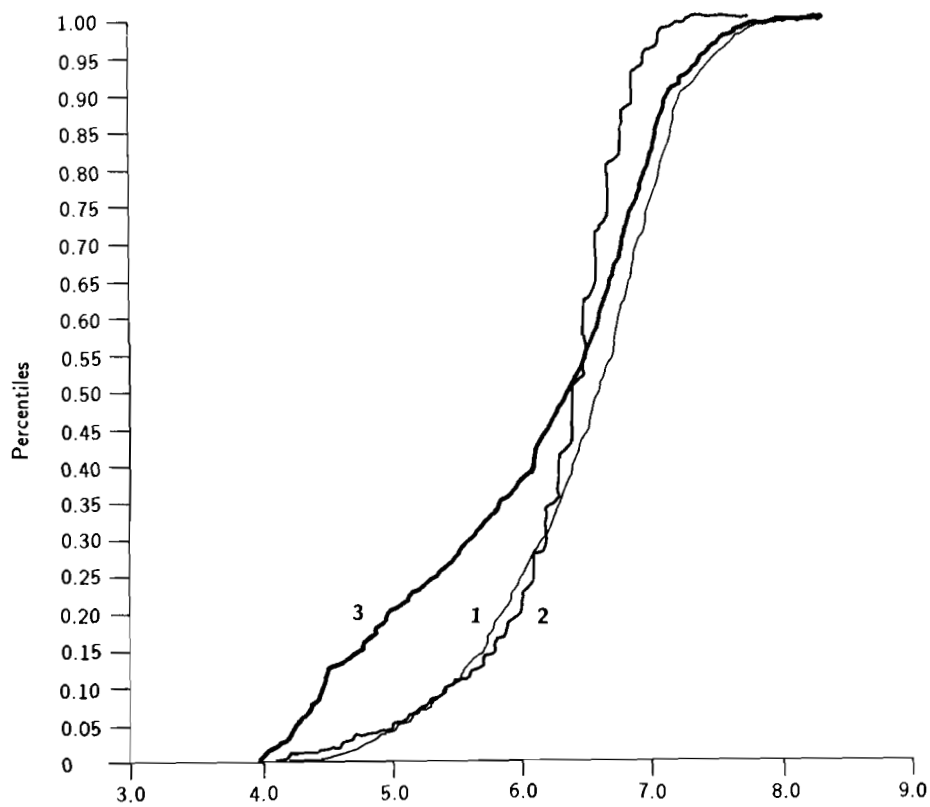

Figure 4.5 Cumulative distribution of the measured pH in 1980 in region 1 of Finland (graph 1), of the RAINS Lake Module response of pH in 1920 (graph 2) and in 1980 (graph 3) as result of 500 Monte Carlo simulations (adapted from Hettelingh and Gardner, 1988).

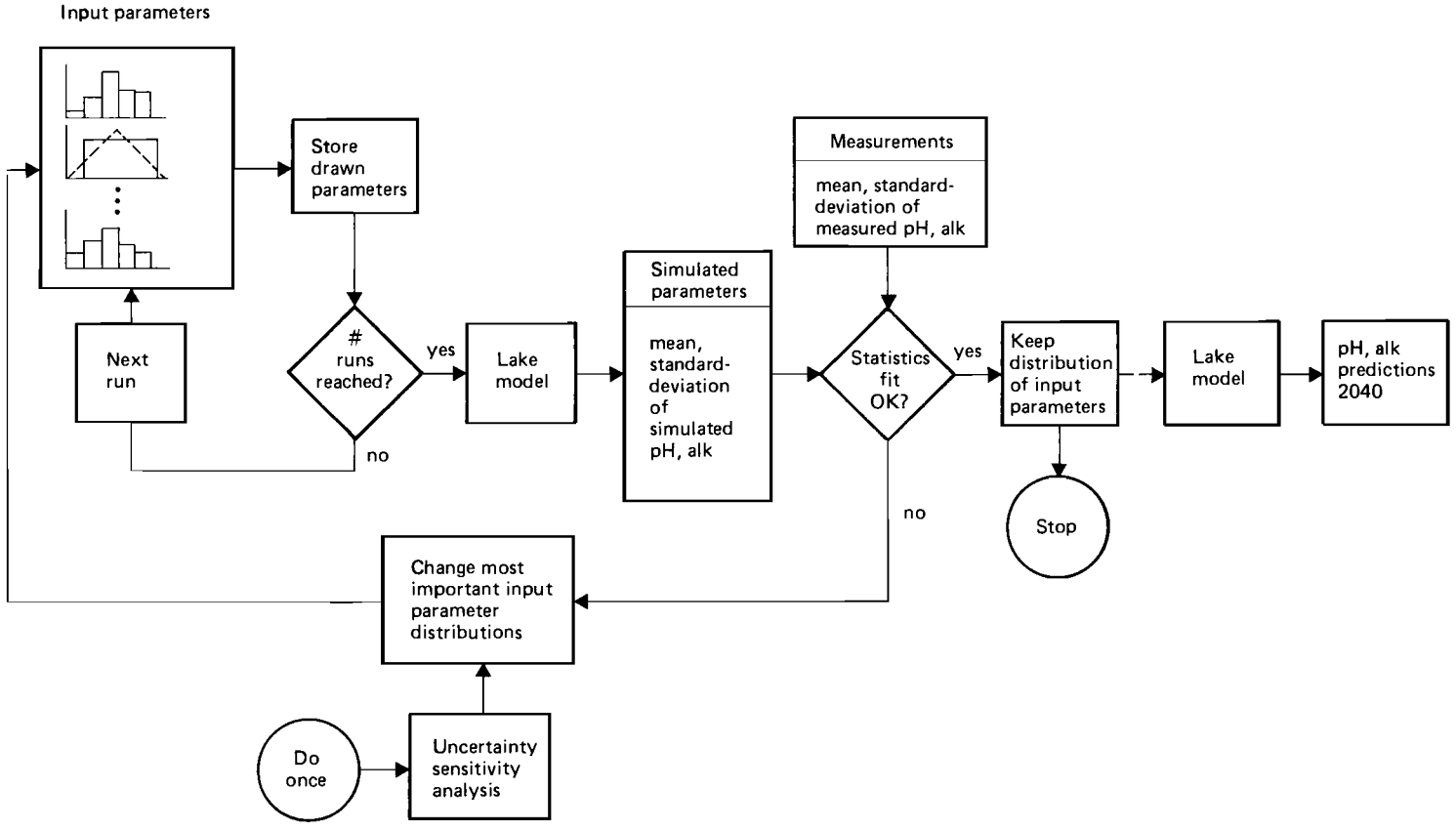

Figure 4.6 The calibration procedure of the RAINS Lake Module by means of PRISM.

Table 4.3 Parameter uncertainty analysis for Monte Carlo simulations of the regions 1 of Finland, Sweden and Norway[1]

Parameter	Finland		Norway		Sweden	
	Rank	%	Rank	%	Rank	%
soilt	1	32.4	1	49.1	1	25.9
sibr	2	7.3	2	16.8	2	9.1
clrat	3	4.8	3	8.3	6	2.4
cond					3	4.8
fcap	4	3.4				
bsata			4	3.2		
cec			5	3.1		
areal	5	2.9	6	2.4	8	1.8
T(12)					5	3.2
P(7)	6	2.9				
ldept	7	2.6				
T(11)			7	1.8		
sy					7	2.0
slope	8	1.9				
evs			8	1.5		
bsatb	9	1.2			4	4.5
T(6)			9	1.5	10	1.1
P(4)					9	1.5
T(8)	10	1.1				
P(12)			10	1.1		

[1]Parameter ranks are based on the percent effect that each parameter has on predicted pH values in 1980. The percent effect is estimated using the partial R^2 statistic (equation 2.16). Values for the uncertainty analysis are determined by simultaneous variation of parameters from pre-specified frequency distributions (see section 2.3 and 2.4). Parameters explaining less than 1% of the pH variation, are not ranked.

If, for example, calibration leads to a Figure like 4.5 in which, however, the difference between the cumulative pH distributions occurs in pH ranges above 6.5, the relevance of this mismatch is not significant from a water-quality standpoint (see Figure 1.2). A combination of the Kolmogorov-Smirnov test and the requirement of a good fit in the lower percentiles would be a possible approach (see Hettelingh and Gardner, 1988). An alternative is to restrict the analysis to the moments and to the tails of the distribution.

In this chapter a calibration method is applied that allows extrapolation of results to other regions and that allows for the estimation of errors and uncertainties of the regionalization. The comparisons of interest for this study are the differences between the

RAINS Lake Module and the Scandinavian data for the mean, variance and the range of predicted pH values. For most applications in environmental modeling a knowledge of the mean and the variances is sufficient (see also Tiwari and Hobbie, 1976). A method first applied by Bartell (1986) simultaneously compares relative biases and the ratio of variances as follows:

$$rB = (\bar{X}_p - \bar{X}_o)/S_o \quad (4.1)$$

$$F = (S_p)^2/(S_o)^2 \quad (4.2)$$

where,

rB = relative bias

$\bar{X}_p$ = mean of the pH model predictions

$\bar{X}_o$ = mean of the pH measurements

S_o = standard deviation of the pH measurements

S_p = standard deviation of the pH model predictions

F = ratio of variances

Relative bias measures model accuracy by quantifying the mean difference between the model and the measurements in units of standard deviations of the data. An exact match leads to an rB of 0. The ratio of variances provides an indication of the relative spread of the model predictions compared to the measurements. If $F < 1$ the model response has a narrower distribution than the measurements while for $F > 1$ the opposite is true. The objective of the applied calibration procedure is:

1. reducing rB to a minimum close to 0.
2. having F converge to 1.
3. having the difference in the ranges of the predicted and measured pH values be as small as possible.

The procedure consists of several iterations of 500 Monte Carlo simulations of the model over the period 1960 to 1980 until the 3 criteria, specified above, are satisfied. For each repetition of the procedure the distributions of the parameters that rank highest (Table 4.3), i.e soilt and sibr are adjusted. After every set of 500 Monte Carlo runs, the rB, F and the percentage of responses outside the

allowable range, were compared to the results of the previous set of runs and to the 3 targets specified above. A new repetition with another shape of the initial distribution or other moments of the most important parameters[13] was started when a previous run had led to better results. The decision whether a run leads to better results was subjectively made, i.e. not all three criteria of a run needed to be simultaneously better than a previous one. For example a slightly higher rB was accepted if F and the percentage outside the range were improved. Table 4.4 lists the extent to which the 3 criteria were met in every calibration iteration applied for the three southern regions of Finland, Sweden and Norway.

Table 4.4 is a result of the adaptation of the shapes and moments of the average soil thickness and the silicate buffer rate. The initial distribution of the average soil thickness (soilt) was iteratively adjusted resulting in a final distribution that was triangular. The procedure involved the simultaneous adaptation of the moments of sibr but not its initial histogram shape.

The assumptions about the shape of soilt does not imply that its actual distribution in a region is triangular, but rather that the effective soil thickness and silicate buffer rate must jointly show a strong central tendency in order to make the mean and the variance of model predictions match those of the measurements. Conversely it is also true that the fact that the other parameters were unimportant, and therefore not adjusted during the calibration process, does not imply that they have been correctly estimated. Thus, parameters are not unique for each region, but represent a set of values which best characterize the model behavior in a particular region.

Table 4.4 illustrates that the values of rB and F in the uncalibrated case, iteration 1, of the southern region of Finland was corrected by the final calibration result of iteration 6. The frequency of model response outside the measured pH range of 1980 was decreased from 8% before the calibration to 7% afterwards. The results show that the southern regions of Sweden and Norway were more difficult to calibrate.

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[13] These changes were made in the file data.dat of PRISM1 (see figure 4.2).

Table 4.4[1] Iterative calibrated pH model response

Country	iteration #	$\bar{X}_p$	S_p	rB	F	% of response outside range
Finland	1	6.42	0.97	0.21	1.27	8
	2	6.28	1.02	0.06	1.38	10
	3	6.36	0.99	0.14	1.32	8
	4	6.38	0.94	0.17	1.19	8
	5	6.44	0.80	0.24	0.86	5
	6	6.30	0.93	0.07	1.17	7
Sweden	1	6.29	1.02	0.57	1.03	11
	2	6.37	0.96	0.64	0.93	11
	3	6.58	0.83	0.86	0.69	7
	4	6.34	0.96	0.62	0.91	11
	5	6.55	0.93	0.82	0.86	6
Norway	1	6.37	0.96	3.19	4.67	0
	2	6.44	0.89	3.35	4.07	0
	3	6.37	0.96	3.19	4.67	0
	4	5.00	0.73	0.08	2.74	0
	5	4.98	0.76	0.04	2.97	0

[1] See equation 4.1 and 4.2 for the meaning of the variables.

The difficulty in calibrating Sweden and Norway may be due to the relatively high deposition (Figure 4.7) in calibration year 1980 as compared to the southern part of finland, and differences in geochemical processes which are not accounted for by the parameters of the RAINS Lake Model. Note, also from Figure 4.7, that the deposition gradient in region 1 of Norway is much less homogeneous than in the first region of Sweden and Finland.

4.3 RAINS LAKE MODEL RESPONSE AS FUNCTION OF PREDEFINED REGIONS

The procedure designed to investigate the effect of regionalization consists of investigating the sources of error which affect regional model prediction. The sources of errors that affect model predictions have been partitioned (see Figure 2.13) in many ways (O'Neill and Gardner, 1979; Gardner et al., 1980b), but the effects on regional environmental models have not been clearly specified.

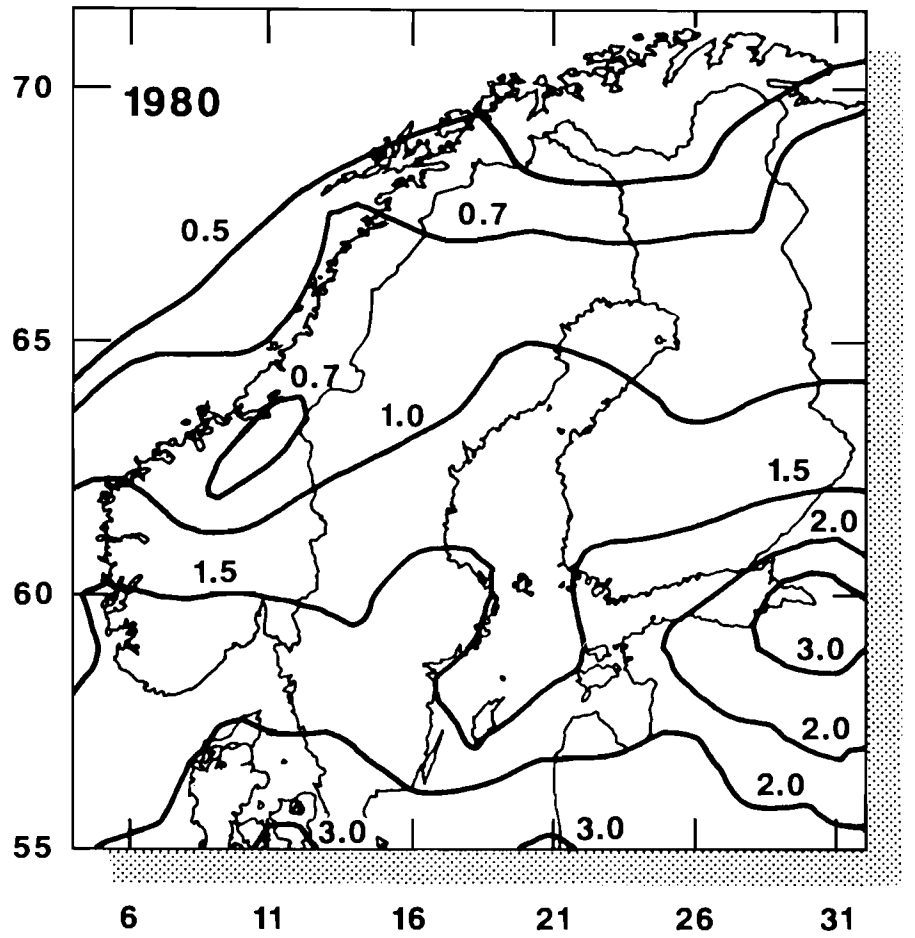

Figure 4.7 The deposition pattern over Scandinavia in 1980, similar for all RAINS scenarios.

In the following three sources of error are assumed to be relevant (Gardner et al., 1987):

1. errors (e_1) due to uncertainties associated with estimates of model parameters,
2. errors (e_2) due to the application of a model on different regions, i.e. when model predictions are extrapolated from one region to another, and
3. residual errors (e_3) due to assumptions and simplifications in the structure and computer implementation of the model.

In general the effects of these errors individually are difficult to distinguish. Residual errors (e_3) and extrapolation errors (e2) will be dependent on the adequacy of parameter estimates. It is, however, important to characterize the errors and, where possible, quantify them in order to verify model applications on regions of environmental subsystems.

The effect of parameter uncertainties (e_1) and residual model errors (e_3) was characterized by superimposing the calibration results (see section 4.2) of the most influential parameters (soilt and sibr) of region 1 of each country on all other regions within the same country. This extrapolation with its associated errors was made for all but the first region within each country and is referred to as the intra-country comparison with an associated intra-country error. Additional errors due to the extrapolation process (e_2) were investigated by superimposing the results of the calibration process and all other input parameters (see Table 4.2), i.e the geomorphologic characteristics, from the first region in Finland to all other regions in Scandinavia. Region 1 of Finland was chosen for this extrapolation because of the good agreement between the model and data (see Table 4.4). Such extrapolation will be called inter country extrapolation with an associated inter country error. Inter country extrapolation assumes that all other regions "behave" as region 1 of Finland except for the pattern and quantity of sulfur deposition.

Intra-country extrapolation

The intra country comparisons are summarized in Table 4.5. The results in Table 4.5 show that in general the estimation of the mean lake pH is not too distant from the measured mean. In Finland the largest deviation, d, of 7.9% (% column) occurs in region 2. The largest deviation in Sweden, 14.5%, occurs in region 1, due to the unsatisfactory calibration. In Norway the prediction of the mean is equally reasonable, with a largest error of 5.5% in the northern most region 3. Note however that the application of the calibration result in the northern part of Finland, starting from region 2, and all northern regions of Sweden result in a reduction of the prediction bias to resp. 2.1% and 0.1% . However, in Norway an inverse pattern is displayed, increasing from 0.4 to 5.5. The effect of a calibration error (e_1) should be expected to show an equal pattern in all intra-country comparisons. A possible cause of the inconsistency may be the joint effect of the calibration error, and the different deposition patterns within the regions. Indeed, from Figure 4.7 it is clear that the heterogeneity of deposition patterns is largest in Norway, in particular in region 1.

The value of d for the first regions of Finland, Sweden and Norway is 0.06, 0.83 and 0.02 with an average for these regions of 0.30. The average value for d for all comparisons is 0.29, indicating that the extrapolation process produces no increase in absolute bias if all 14 regions are investigated in an aggregate way.

The values of rB are all less than 1.0, indicating that the mean of the model predictions are always less than one standard deviation from the data. The ratio of the variances, F, is less than 1.0 in the regions 2 through 5 of Finland, near 1.0 in the regions 2 through 6 of Sweden, and above 1 in the regions 2 and 3 of Norway. The combination of rB and F statistics is displayed in Figure 4.8. Ideally, model predictions should result in (rB,F)-clusters around (0,1). The underestimation of the variability of the predictions in all the regions of Finland, most of the regions in Sweden, and its overestimation in all the regions of Norway demonstrate that the calibration error in the regions will affect the fit between the

extreme percentiles of the cumulative distributions of the predictions and those of the measurements.

Table 4.5[1] Intra-Country Comparisons of 1980 Measured pH Values Against Model Predictions[2]

Region	DATA		MODEL		d	%	rB	F
	$\bar{X}^o$	CV	$\bar{X}^p$	CV				
Finland								
1	6.24	13.8	6.30	14.8	0.06	1.0	0.07	1.15
2	6.31	11.3	6.81	8.4	0.50	7.9	0.70	0.63
3	6.47	12.1	6.83	9.1	0.37	5.7	0.47	0.64
4	6.58	11.6	6.94	8.1	0.36	5.5	0.48	0.53
5	6.79	9.4	6.93	5.9	0.14	2.1	0.21	0.40
sum/5					0.29			
Sweden								
1	5.72	17.5	6.55	14.2	0.83	14.5	0.82	0.86
2	5.30	22.1	5.84	17.5	0.54	10.2	0.46	0.76
3	6.33	18.3	6.26	14.7	0.07	1.1	-0.06	0.62
4	5.84	12.3	6.41	12.9	0.58	9.9	0.80	1.33
5	6.36	11.0	6.51	11.8	0.14	2.3	0.21	1.19
6	6.61	12.1	6.60	9.5	0.01	0.1	-0.01	0.62
sum/6					0.36			
Norway								
1	4.96	8.9	4.98	15.3	0.02	0.4	0.04	2.97
2	5.54	15.2	5.48	15.7	0.07	1.3	-0.08	1.03
3	5.96	8.9	6.29	11.6	0.33	5.5	0.61	1.88
sum/3					0.14			
sum/14					0.29			

[1] adapted from Gardner et al., 1987. [2]The CV is the relative variability calculated as the (standard deviation / mean) X 100. d is the absolute value of the difference between the mean of the data and the mean of the model. The mean absolute value of d for each country and for the entire table are also listed. The % column gives d as a percentage of the mean of the measurements.

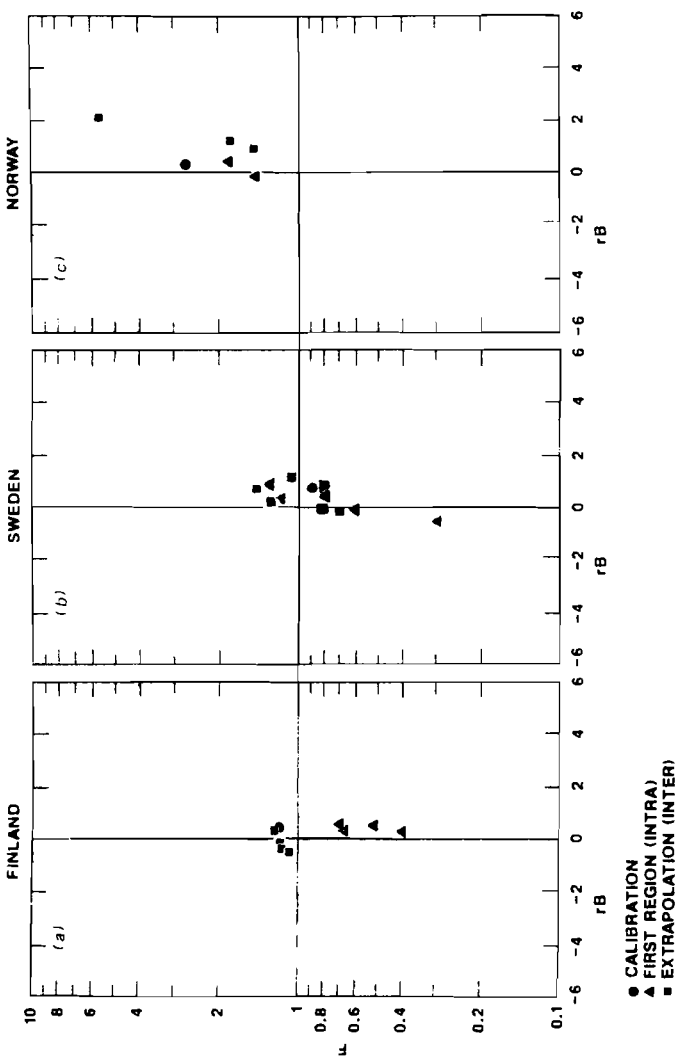

Figure 4.8 Relative bias (rB) and ratio of variances (F) resulting from calibration, intra extrapolations and inter extrapolations (adapted from Gardner et al., 1987)

Inter-country extrapolation

Table 4.6 summarizes the results of the inter country comparisons of the RAINS Lake Model simulations compared to available measurements. The inter country comparisons are based on the extrapolation of all parameters from region 1 of Finland (Figure 4.1) to other regions within Scandinavia, except for the deposition. For every region the prevailing sulfur deposition ranges (Figure 4.7) is applied.

The absolute deviation (d) is smallest in Finland and largest in Norway. The average over all Scandinavian regions of the absolute deviation d (0.39) has increased compared to the result of the inter country extrapolation (0.29, Table 4.5). The average over the regions 1 in the three countries has also increased from 0.30 (Table 4.5) to 0.58 (Table 4.6) indicating that the calibration and regional extrapolation error jointly (e_1 U e_2) lead to a larger error, than the average absolute deviation over all regions (0.39 in Table 4.6); aggregation of regions smoothes the error.

The inter-country extrapolation for Finland is generally better than the intra-country (Table 4.5) comparison (recall that only the soil thickness (soilt) and the silicate buffer rate (sibr) were extrapolated in the intra-country comparison). The relative bias of results, rB, is less and the ratio of the variances, F, more closely estimates the variability of the data. Figure 4.8 shows that the (rB,F) cluster of the inter country extrapolation in Finland is closer to (0,1).

The results of the inter-country extrapolation for Sweden are not consistently higher or lower. Except for region 2 and, to a lesser extent, region 6 the absolute deviation d is lower than in Table 4.5. The difficulties in predicting the regional behavior of lakes in region 2 may be due to interaction of high rates of sulfur deposition, a relatively strong deposition gradient differences (see Figure 4.7) and differences in geochemical processes which were not accounted for by the parameters of the Rains Lake Model. The joint effect of the calibration (e_1) and regional error (e_2) thus increases the prediction error in region 2. However, the average over the Swedish regions of d in the case of the inter-country comparisons (0.35, Table 4.6) is similar to the average of d in the case of the intra-country comparisons (0.36, Table 4.5). This suggests that the prediction errors

in the intra and inter comparisons, become less distinguishable when the region is heterogeneous with respect to deposition levels, calibration and regional error.

The inter-country extrapolations are rather poor for the three regions in Norway.

Table 4.6[1] Inter-Country Comparisons of 1980 Measured pH Values Against Model Predictions[2]

Region	DATA		MODEL		d	%	rB	F
	$\bar{X}_o$	CV	$\bar{X}_p$	CV				
Finland								
1	6.24	13.8	6.30	14.8	0.06	1.0	0.07	1.15
2	6.31	11.3	6.41	13.1	0.10	1.6	0.14	1.37
3	6.47	12.0	6.39	13.3	0.08	1.2	-0.10	1.20
4	6.58	11.6	6.43	12.7	0.15	2.2	-0.19	1.14
5	6.79	9.4	6.55	10.7	0.24	3.5	-0.37	1.21
sum/5					0.13			
Sweden								
1	5.72	17.5	6.15	17.1	0.43	7.5	0.43	1.10
2	5.30	22.1	6.18	16.7	0.89	16.8	0.76	0.77
3	6.33	18.3	6.25	16.6	0.08	1.2	-0.07	0.71
4	5.84	12.3	6.37	13.6	0.53	9.2	0.74	1.45
5	6.36	11.0	6.44	12.6	0.08	1.2	0.11	1.33
6	6.61	12.1	6.54	11.0	0.07	1.1	-0.09	0.81
sum/6					0.35			
Norway								
1	4.96	8.9	6.22	16.1	1.26	25.4	2.85	5.08
2	5.54	15.2	6.44	12.5	0.90	16.2	1.07	0.91
3	5.96	8.9	6.56	10.7	0.60	10.1	1.12	1.72
sum/3					0.92			
sum/14					0.39			

[1]adapted from Gardner et al., 1987.

2The CV is the relative variability calculated as the (standard deviation / mean) X 100. d is the absolute value of the difference between the mean of the data and the mean of the model. The mean absolute value of d for each country and for the entire table are also listed. The % column give d as a percentage of the mean of the data.

Both the absolute (d) and relative (rB) bias are high and the variance is poorly estimated for region 1 (F = 5.08, Table 4.6). In Figure 4.8 the (rB,F) cluster of Norway is shown to be distant from (0,1).
The prediction error in the three regions in Norway is clearly affected by the regional characteristics, i.e. corresponding to region 1 of Finland, that are imposed. The variances of the prediction have increased in comparison to the intra-extrapolation, in almost all regions, thus affecting the confidence that may be attributed to the tails of the cumulative distribution of model response.

Time dependent extrapolation

Model simulations of the mean pH values in the year 2040 for two sulfur deposition scenarios were performed with the intra- and inter-country parameter sets. Because these predictions can not be verified by data, the results will concentrate on the differences due to the interaction of the two extrapolation procedures with the sulfur deposition scenario (Figures 4.3 and 4.4). This difference, D, between the intra- and inter-country simulations for each scenario is given in Table 4.7.

The time extrapolation of the calibration and regional error tends to increase the average prediction error when deposition accumulates over time in Finland (from 0.34 to 0.41, 0.34), in Sweden (from 0.15 to 0.17, 0.18) and in Norway (from 0.82 to 0.97, 1.03). This increase is however not similar in all countries. The dominating calibration error ($\mathbf{e}_1$) in Finland (underestimation of intra-variances; Figure 4.8) remains constant under the high deposition. However, from comparing the high deposition pattern in 2040 (Figure 4.3) with the deposition pattern in 1980 (Figure 4.7) it can be seen that hardly any difference occurs in Finland. The low deposition pattern in 2040 (Figure 4.4), on the contrary, is different from the 1980 pattern.

The average over the Finnish regions of the prediction errors consequently becomes larger (from 0.34 in 1980 to 0.41 in 2040). The dominating regional error ($\mathbf{e}_1 \cup \mathbf{e}_2$) in Norway (overestimation of inter-variances; Figure 4.8) leads to an important increase of the prediction error in both the low and high deposition case.

Table 4.7[1] Differences Between Inter and Intra Country Comparisons for the High and Low Sulfur Deposition Scenarios[2]

Country Region	1980		Scenario[2] for Year 2040			
			Low		High	
	D	%	D	%	D	%
Finland						
1	0.00	0.0	0.00	0.0	0.00	0.0
2	0.40	6.3	0.40	6.2	0.41	6.1
3	0.44	6.5	0.43	6.2	0.48	7.1
4	0.51	7.5	0.84	6.8	0.49	7.1
5	0.38	5.5	0.37	5.3	0.36	5.2
sum/5	0.34		0.41		0.34	
Sweden						
1	0.40	6.1	0.42	6.3	0.44	6.9
2	0.34	5.8	0.35	5.9	0.42	7.6
3	0.01	0.2	0.05	0.8	0.08	1.3
4	0.04	0.6	0.02	0.3	0.02	0.3
5	0.07	1.1	0.12	1.8	0.05	0.8
6	0.06	0.9	0.14	2.1	0.03	0.5
sum/6	0.15		0.18		0.17	
Norway						
1	1.24	25.0	1.58	33.3	1.43	31.1
2	0.96	17.5	1.13	20.9	1.34	26.7
3	0.27	4.3	0.19	2.9	0.31	5.0
sum/3	0.82		0.97		1.03	
sum/14	0.37		0.43		0.42	

[1] adapted from Gardner et al., 1987

[2] D is the absolute value of the difference between sets of simulations calibrated to the first region of each country (Table 4.5) and sets of simulations calibrated to the first region of Finland and extrapolated to other countries (Table 4.6). The % column gives the values of D as a percentage of the mean predicted value of Table 4.5. The mean absolute value of D for each country and for the entire table are also listed.

In Sweden where the calibration as well as the regional error lead to a somewhat similar predicting error (cluster around (1,0) in Figure 4.8) no major change of the prediction error over time occurs.

The difficulties of calibrating and predicting lake pH for regions 1 and 2 in Sweden are still evident by inspection of D in 2040. The extrapolation errors made in Norway (Table 4.6) also lead to an increase D from 1980 to 2040, especially if the deposition is high.

The mean predicted pH in 2040 are shown in Figure 4.9 for the calibration regions (solid circles), intra-country extrapolations (solid triangles) and inter-country extrapolations (solid squares). The simulations of the high sulfur deposition scenario and low deposition scenario are connected by a solid line, with the high deposition scenario always predicting a lower mean pH. Each point illustrated in Figure 4.9 represents 500 independent Monte Carlo simulations. On average the mean difference between scenarios ranges from 0.18 to 0.28 pH units.

In summary

The results indicate that (1) if the RAINS Lake Module is used as a measure of D (the net difference in pH due to the different sulfur deposition scenarios), then its calibration and regional error are less important. (2) If, however, the accuracy of the pH simulations is more important, then , as has been shown from the intra and inter country comparisons, the importance of regional and calibration error may affect the confidence of the prediction of watershed quality in a region. Differences in geomorphological conditions and deposition patterns have been shown to interact in a spurious way, thus affecting regional as well as temporal predictions that are based on calibration results.

Regional and temporal predictions are evaluated in the next section when in stead of the RAINS Lake Model another more aggregated model structure, i.e a metamodel is used.

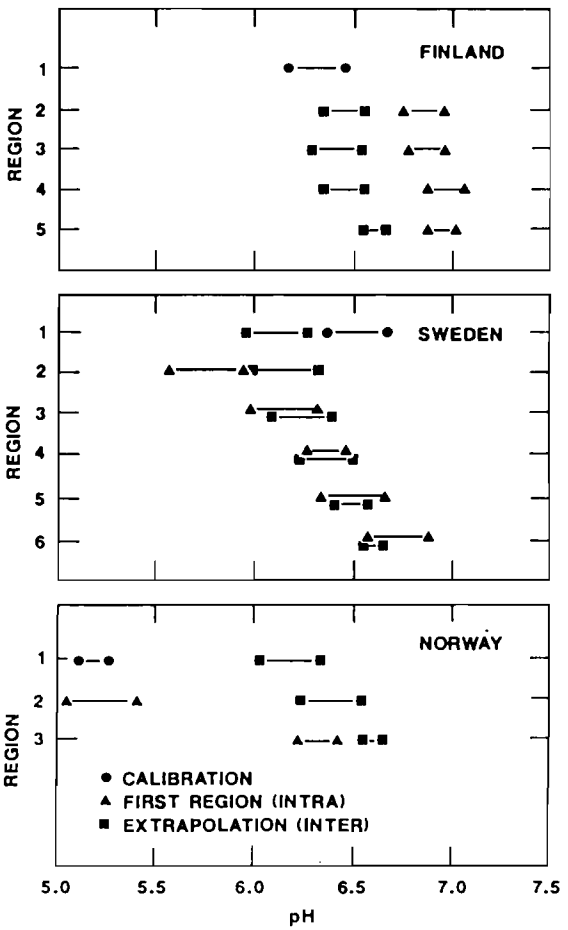

Figure 4.9 Comparison of mean pH model predictions in 2040 for the Low (left squares and rectangulars) and for the High Scenarios. (adapted from Gardner et al., 1987)

4.4 METAMODEL RESPONSE AS FUNCTION OF PREDEFINED REGIONS

In this section the quality of the information on regional characteristics is investigated, that must be met in order to make pH predictions as function of deposition patterns and calibration. For policy purposes it is important to be able to make predictions also when information on geomorphological conditions is lacking and/or varying in a predefined region.

Therefore, the aim is (1) to investigate whether the information contained in the application of a complex model in a region can be condensed to a small subset of information and (2) to investigate what the influence of model complexity is with respect to the issue of model predictions in fixed regions, treated in section 4.3.

Region and deposition pattern

Region 1 of Finland was chosen, because of its relatively good coverage in the sample (Table 4.2), in combination with deposition ranges in 1980 resulting from the Official Energy Pathways (see Figure 4.7). The simulation period chosen for the analysis was arbitrarily set three times as long as the standard calibration period within RAINS, i.e. sixty years covering 1920 to 1980. The reason for a long period is to reduce the effect of uncertainties associated with the initial values for starting the model simulations. The deposition values for the years before 1960 were obtained by disaggregating the historical emission totals (Fjeld, 1976, cited in Whelpdale, 1987) over the European countries on the basis of the 1980 emissions per country, and applying the RAINS deposition module (chapter 2). The minimum and maximum deposition in Finland region 1 ranged in every year within the interval between 0.5 $g/m^2/yr$ and 2.0 $g/m^2/yr$.

A four step procedure was designed as follows:

1. Identify important parameters: An uncertainty analysis similar to the one conducted in section 4.3 was performed, using 60 deposition input ranges (sy, Table 4.2). The result is presented in Table 4.9.

The ranking of Table 4.9 is different from that in Table 4.3. This

indicates that different deposition patterns, on a region, can lead to a shift in uncertainties of model parameters.

2. designing a metamodel: The purpose of the second step is to use the important parameters of step 1 along with appropriate deposition parameters to form an empirical relationship that can be used to predict the response of the model in 1980. It is reasonable to assume that acidification is a function of acid deposition in a current and past years. This process of the lagged influence of acid deposition is well taken into account by the recursiveness of the RAINS Lake Module (chapter 3).

Table 4.9[1] RAINS Lake module parameters explaining more than 1%[2] of the 1980 pH response.

Parameter[3] Name	Rank	%[2]
soilt	1	54.8
calk	2	6.0
sibr	3	4.0
pfac	4	1.2
cec	5	0.8

[1] Adapted from Hettelingh and Gardner, 1988.
[2] % contribution of parameters to the variance of the pH response in 1980 is computed by means of the partial R^2 statistic (chapter 2).
[3] parameter names are explained in Table 4.1.

For this reason the pH response of the metamodel in 1980 (see for a description of metamodeling chapter 1 and chapter 2) has been assumed a function of accumulated sulfur deposition in the period 1920-1980.

The inclusion of deposition as explanatory variable, in spite of its relative non-importance (Table 4.9), is a result of the apparent relation between deposition and regional characteristics of section 4.3.

3. calibrating the metamodel: Calibration of the regression equation was accomplished by performing Monte Carlo simulation on the equation and comparing the cumulative distribution of the metamodel response against the cumulative distribution of the measured pH in 1980. The calibration convergence criterion consists of the Kolmogorov-Smirnov test in conjunction with an acceptable fit of the lower percentiles (see also section 4.2).

4. reducing the number of explanatory variables of the metamodel: this step is introduced to increase the applicability of the metamodel in a region where even not enough information is available to use the equation of step 3. Another reason is that the generality of the method proposed here is unnecessarily affected by the adjustment of the input parameter distributions as part of the calibration procedure in step 3. The ability to obtain a metamodel with unchanged measured (input) parameter distributions evidently increases the application of such a simple model on other regions.

Response surface models, including metamodels, can be formulated using statistical techniques (regression, principal components) or be based on intuition. In either case will the fit of the relationship be improved by the calibration procedure of step 3 in accordance with step 4. Following Kleijnen's (1987) application of regression techniques in the development of metamodels, ordinary least squares (OLS) is applied in step 2.

Different metamodels were tested. A suitable least squares relationship, defined in step 2 was found to be equation 4.3 :

$$\ln(SpH_{1980}) = \underset{(0.008)}{0.13}*\ln(soilt) + \underset{(0.009)}{0.06}*\ln(sibrc) + \underset{(0.003)}{0.02}*\ln(cec) + \underset{(0.005)}{-0.02}*\ln(calk) \underset{(0.003)}{- 0.004}\ln(pfac) \underset{(0.0006)}{- 0.003}*Sdep + \underset{(0.07)}{0.62}*\ln(SpH_{1920}) \quad (4.3)$$

where,

SpH_{1980} = pH model response of the RAINS Lake Module in 1980

SpH_{1920} = pH model response of the RAINS Lake Module in 1920

Sdep = $\ln(sy_{1920}) + \ln(sy_{1921}) + \ldots + \ln(sy_{1980})$

(see Table 4.1 for the meaning of the other parameters)

The numbers between brackets indicate the standard deviations of the estimated coefficients. Figure 4.10 displays the cumulative

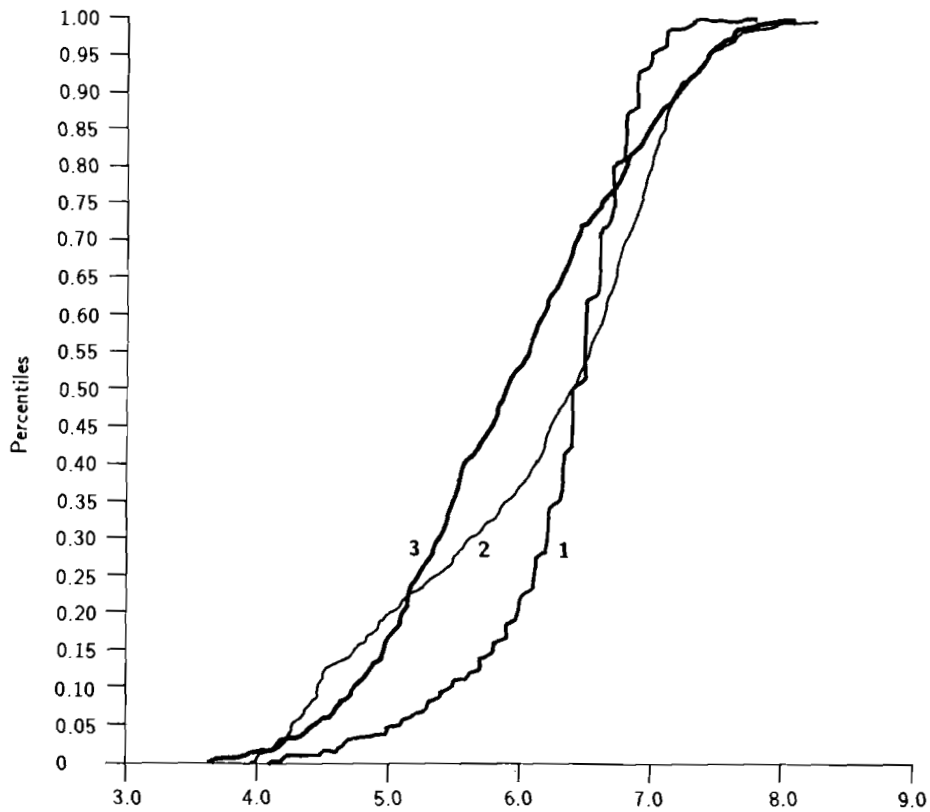

Figure 4.10 Cumulative distribution of the measured pH in 1980 in region 1 of Finland (graph 1), of the RAINS Lake Module response (graph 2) and of metamodel 4.1 (graph 3) of the pH in 1980 as result of 500 Monte carlo simulations (adapted from Hettelingh and Gardner, 1988)

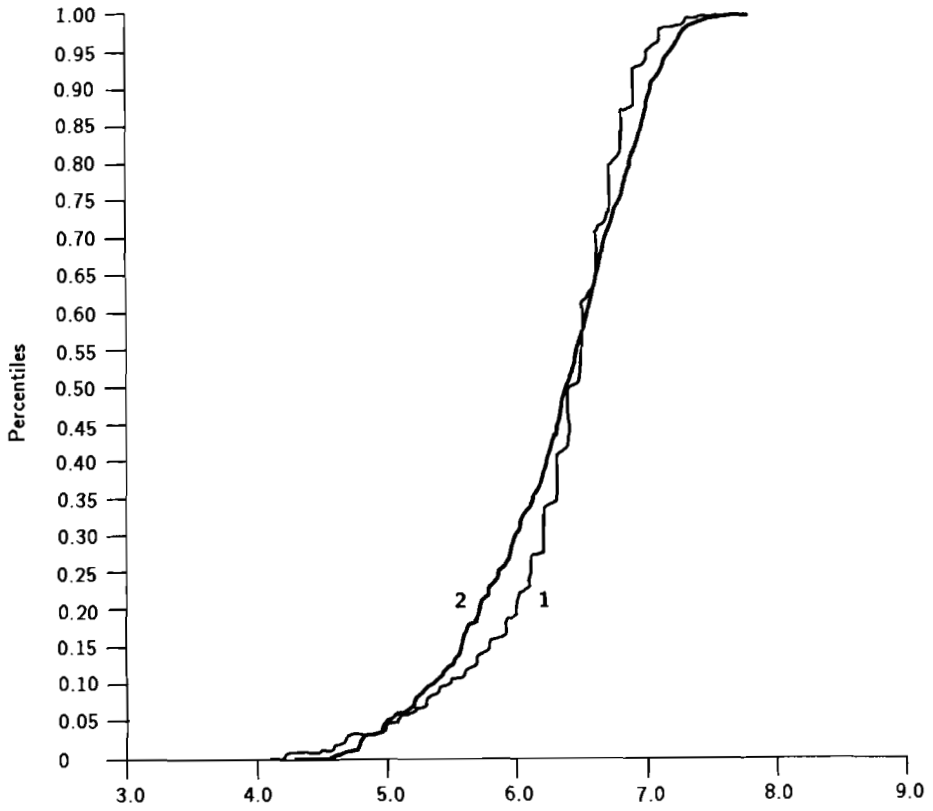

Figure 4.11 Cumulative distribution of the measured pH in 1980 in region 1 of Finland (graph 1), and of the calibrated response of metamodel 4.1 (graph 2) as result of 500 Monte Carlo simulations (adapted from Hettelingh and Gardner, 1988).

distribution of the simulated pH from equation 4.3 (graph 3)[14], the model response of the RAINS Lake Module (graph 2) and the pH measurements in Finland region 1 (graph 1). Both simulation results deviate substantially from the 1980 pH measurements. The metamodel performs better than the RAINS Lake Module in the lower but worse in the higher percentiles.

Monte Carlo simulations were performed (step 3) on equation 4.3 by assuming that the initial values of the lake pH in 1920 lie within the neutral range, i.e. between 6 and 8 with a mean of 7, with the shape of SpH_{1920} assumed to be triangular. By iteratively changing the moments of SpH_{1920} and **soilt**, the difference between the cumulative distribution of the metamodel response and the measurements was gradually reduced. The final result of this calibration is shown in Figure 4.11. A two-sided Kolmogorov-Smirnov test was performed by comparing the predictions of the metamodel (graph 2) with the measurements (graph 1). A statistic of 0.13 was obtained which allows for the rejection of the null hypothesis that the two graphs are similar[15].

It is of more importance for the prediction of water quality to obtain a good fit in the lower pH ranges rather than a reasonable fit over the entire pH range. Therefore, the Kolmogorov-Smirnov test was used as convergence criterion of the calibration procedure. A calibration run was considered better than a previous run if it resulted in a lower Kolmogorov-Smirnov test value, in conjunction with an improvement of the lower percentiles of the simulation result. The absolute deviation of the 2.5, 25 and 50 (median) percentiles of graph 1 and graph 2 as percentage of the similar percentiles in graph 1 are respectively 3, 3 and 0.2%. The application of this combined criterion indicated that no better calibration to the measurements could be obtained.

Entering step 4 of the analysis the question is if all variables of equation 4.3 are relevant from the point of view of being able to apply the equation in another region. The 1% inclusion criterion (Table 4.9),

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[14]The distribution is obtained by performing 500 Monte Carlo simulations on equation 4.1 over the period 1920-1980.

[15]the number of measured pH values in region 1 of Finland equals 819 and the number of Monte carlo simulations equals 500. The significance for the two sample test is 0.05.

for instance, lead to the inclusion of pfac which is nothing more than a weight attributed to precipitation (see equation 3.22), which was relevant for the RAINS Lake Module. Moreover assumptions were made about the shape of SpH_{1920} in order to calibrate equation 4.3, which affects its application for another combination of regional characteristics and deposition patterns. Therefore it is of interest to accommodate another metamodel to the results of equation 4.3. For this purpose the Monte Carlo simulation results of equation 4.3 were regressed on soilt, calk and accumulated deposition only:

$$\ln(MpH_{1980}) = \underset{(0.025)}{0.1306}*\ln(soilt) - \underset{(0.0016)}{0.02}*\ln(calk) + \underset{(0.0009)}{-0.009}*Sdep \qquad (4.4)$$

where,

MpH_{1980} = Monte Carlo simulation response of equation 4.3

A Monte Carlo simulation and analysis of equation 4.4, lead to a cumulative distribution of the predicted pH in 1980 that is not significantly different from the measurements in 1980 at 0.01 significance (statistic of 0.09). In addition the fit is relatively good in both the lower and higher percentiles (Figure 4.12)

Temporal and spatial extrapolations with the metamodel

In order to test spatial and temporal robustness of equation 4.4 two extreme tests were performed. The first test was to apply the metamodel on the most northern part of Finland (region 5, see Figure 4.1) where low deposition levels occur (between 0 and 0.5 $g/m^2/yr$). The second test was to use equation 4.4 with a ten fold higher deposition (between 10 and 20 $g/m^2/yr$) thus simulating in an extreme way a distant future deposition accumulation[16].

[16]Note that deposition levels higher than 10 $g/m^2/yr$ are not uncommon in central Europe.

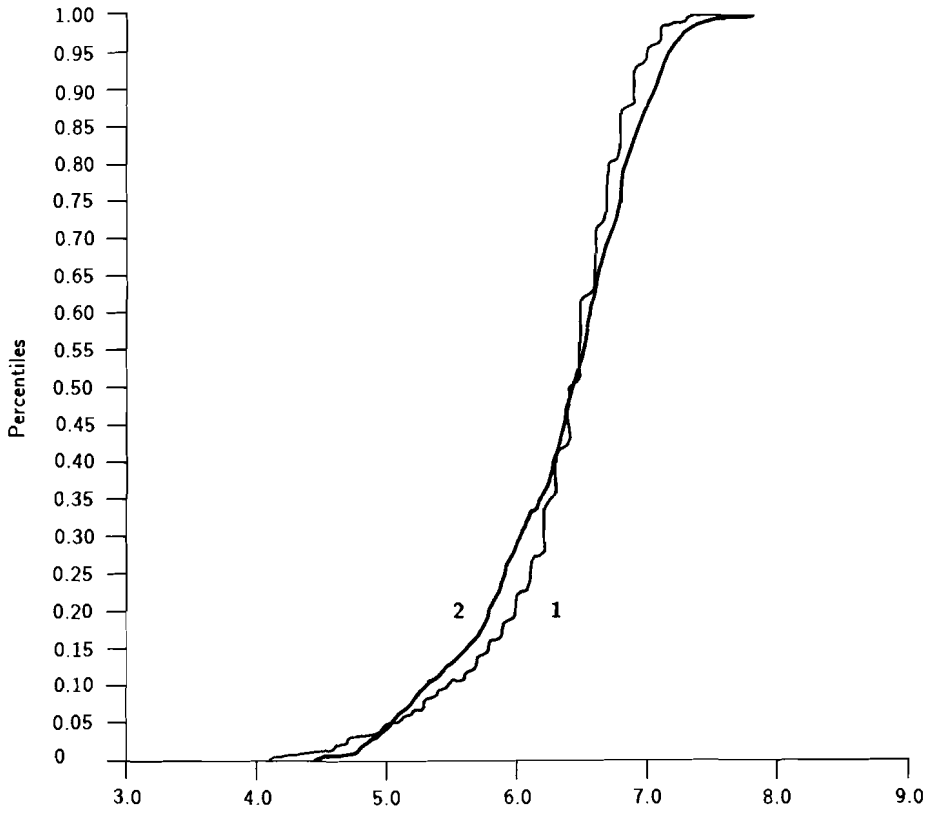

Figure 4.12 Cumulative distribution of the measured pH in 1980 in region 1 of Finland (graph 1), and of the response of metamodel 4.2 (graph 2) as result of 500 Monte Carlo simulations.

Applying equation 4.4 in a Monte Carlo simulation using regional inputs of the Northern part of Finland (region 5), leads to a mean of absolute deviations between the predicted pH values of 1980 and the measured pH values which is as great as 36% of the mean of the measured pH values. Simulations with the RAINS Lake module in region 5 with a deposition pattern resulting from the Official Energy Pathways scenario lead to a mean of absolute deviations that is not greater than 12% of the mean of the measured pH value. The possible reasons for this difference are that a shift in parameter uncertainties occurs in region 5 as compared to region 1. The three most important parameters in region 1 (Table 4.8) are also most relevant in region 5, but explain respectively 43.4%, 21.8% and 6.1% of the variance in the pH response of the RAINS Lake Module.

The application of an extremely high deposition scenario in region 1 leads to a mean of absolute deviations between the metamodel response and the measured pH values of about 70% of the mean of measured pH values whereas a similar deposition input to the RAINS Lake Module results in a mean of absolute deviations of about 22%.

Summarizing it can be remarked that metamodel applications must be restricted to regional and deposition conditions that are similar to the calibration case. In the calibration case a metamodel, that consists of only the most important parameters of the RAINS Lake Model, may be used within a fixed region instead of the RAINS Lake module. When the combination of regional characteristics and deposition levels change the predictions of a metamodel Model are liable of being error prone.

4.5 CONCLUDING REMARKS

The main proposition of this chapter was that the choice of a zoning system may influence the model calibration results and thus affect the quality of model predictions.

The zoning system that was used is similar to the spatial disaggregation applied for the lake acidification module of RAINS, i.e. partitioning Scandinavia into 5 regions in Finland, 6 in Sweden and 3 in Norway. Two kinds of calibration were applied:

(1) fitting the mean, standard deviation and tails of the RAINS Lake Model pH predictions of 1980 to the same statistics of the measurements. The temporal scale was also chosen similar to the time horizon used in RAINS, i.e from 1960 to 2040. This calibration method was applied to the southern regions, i.e. the regions with number 1 (Figure 4.1), of each of the Scandinavian countries. The best calibration result was obtained in region 1 of Finland and the worst in region 1 of Norway.

(2) fitting the cumulative distribution of the pH predictions in 1980 of a metamodel to the cumulative distribution of the measurements. The calibration especially aims at providing a good fit in the lower percentiles of the cumulative distribution of the pH predictions because this pH range is relevant from a water quality point of view. The spatial scale was restricted to the southern part of Finland because of the relatively good sample by which lake characteristics were described. The temporal scale was taken from 1920 to 2040.

The investigation conducted with both the calibration methods concentrated on (a) the quality of the model predictions under different regional conditions (soil characteristics and deposition patterns) from the conditions under which the calibration was performed, i.e regional extrapolation and (b) the quality of future predictions, i.e temporal extrapolation.

With both calibration method (1) and (2) it was shown that changing combinations of deposition levels and regional characteristics may lead to shifts in the ranking and/or the magnitude of the uncertainty of model parameters. This shift in uncertainties leads to a changing variability of the model predictions, thus affecting the quality of the predictions.

The quality of the predictions made with calibration method (1) was tested by applying the calibration result of the southern regions of Scandinavia to the other regions, and consequently, different deposition ranges in the respective countries. This kind of extrapolation within a country was termed intra-country extrapolation, and was aimed at testing the effect of a calibration error. Besides intra-country extrapolation the effect of applying the regional characteristics of the southern part of Finland to all other regions, i.e. inter-country extrapolation was investigated. Inter country

extrapolation aims at testing the effect of a combination of calibration error and errors due to defining wrong regional characteristics.

It was shown that intra-country extrapolation led to prediction errors that were different in one country to another. The variability of the predictions was underestimated in Finland, mostly underestimated in Sweden and overestimated in Norway. The inter-country extrapolation led to varied results. In Finland predictions were better, in Sweden remained comparable to, and in Norway became worse than the prediction due to intra country extrapolation.

Temporal extrapolation showed that the dominating calibration error in Finland leads to a larger prediction error in future years as compared to the calibration year when the deposition pattern changes. This result also occurs in Norway where the regional error is dominating the error in model results. In Sweden where the calibration error cannot be well distinguished from the regional error, the prediction error was shown to remain about the same.

The quality of predictions made with calibration method (2) was tested by applying the calibration result in the Northern part of Finland and by predicting the pH of 1980 in region 1 of Finland when the deposition increases drastically. First it was shown that the 1980 predictions of a metamodel can be calibrated to obtain a cumulative distribution that is not significantly different from the measurements. Second it was shown that, as with calibration method (1), a prediction error occurs when regional conditions and deposition ranges, that are used as input into the model, become different from the combination of conditions under which the model was calibrated.

In general, from both kinds of investigations, it can be concluded that the merit of calibration for policy purposes, in cases where models are applied to predefined zones in a regional environmental analysis may be limited; environmental policy is concerned with the evaluation of environmental effects due to measures leading to different deposition patterns over a large scale. The predictions necessary for such evaluation may become error prone.

This conclusion has to be formulated in a conditional way, because (1) it has not been investigated what the thresholds are at which regional and deposition changes lead to wrong predictions; the number

of predictable states of a system may be large, varying between the state for which the model was calibrated and the state at which the system collapses, due to a sudden impact. For the latter situation, models need to be used that apply catastrophe theory (see also Zwick, 1978) rather than calibration methods. Another reason (2) is that the conclusion is based on results of using two models that are different with respect to complexity. The RAINS Lake model has mechanistic characteristics, i.e it explains chemical soil and lake processes in some detail. However as was pointed in earlier chapters, systems can be interpreted in many ways, depending on holistic or reductionist views on system modeling. Lake acidification models exist that are much more detailed than RAINS e.g. ILWAS (see chapter 3). The extent to which complexity interferes with the result obtained has not been established; it was shown that the results hold for the RAINS Lake Model and a much simpler metamodel.

However, it must be noted that the result is similar to what was found in the field of spatial interaction modeling. Openshaw (1977a, 1977b, 1978, 1983) showed that the zoning system and model predictions were interdependent, with the result that different regional partitioning lead to different relations within and between regions.

In the next chapter an alternative method is proposed, in which a zonal system is used in relation to RAINS Lake Model predictions without the application of calibration.

5. ZONING OF RAINS LAKE MODEL PREDICTIONS

5.1 INTRODUCTION

The application of site specific models to regions is limited by the influence of regional characteristics and deposition patterns on model calibration. The previous chapter has shown that the result of a calibration in a predefined region may not lead to consistent predictions in other regions. This lack of prediction confidence also occurs when in a predefined region a calibrated model is used to predict effects of deposition patterns that differ from the calibration case. The reason is that a study area can be partitioned in a virtually infinite number of ways, each of which would combine with deposition patterns to different model calibration results.

In spatial interaction modeling, the uncertainty concerning the identification of the objects of spatial study, known as the modifiable-areal-unit problem (Openshaw, 1978; Openshaw and Taylor, 1983), has also been recognized. In this discipline alternative methods to choose a zoning system were developed. Openshaw and Taylor (1983) distinguish (1) filtering methods to identify trends of real patterns by removing noise caused by aggregation effects (2) arbitrary design criteria in which zones are chosen to match particular problem descriptions (3) information theoretic methods determining how many zones are needed to describe a particular phenomenon (4) statistical methods implying that the methods of sampling and zoning are analogous (5) traditional methods aimed at defining zones that have a geographical rationale and (6) optimal zoning methods in which zones are chosen as function of the results that are desired. Openshaw and Taylor (1983, p. 67) describe the optimal zoning method as "... a methodology for formulating and testing spatial hypotheses by deliberately and purposefully exploiting the uncertainty that exists in zonal data because of the modifiable areal unit problem."

The modifiable areal unit problem has not been solved in the sense that a unique method has been found which is always applicable. Openshaw and Taylor (1983, p.67) argue that "... it is necessary for

geographers to be more aware of the geography of the methods they employ and the zoning systems they study. In the longer term it would be nice if the possibilities opened up by the optimal zoning approach for a more geographical form of geographical analysis could be realized."

Undoubtedly, there exists some analogy with the problem described in chapter 4, i.e. both the environmental and geographical model applications lead to results that are dependent on the chosen zonal system. The difference is that the spatial interaction models describe objects that have intra zonal or inter zonal relations. In watershed quality modeling this kind of relationships do not occur. A zone may contain different classes of lake types, each of which reacts in a particular way, i.e. delayed or fast, to acid deposition. But, unless a lake is connected to another, the decrease of water quality of a watershed will not depend on another lake.

This chapter introduces an alternative concept to model calibration in fixed regions, aimed at increasing prediction reliability. The basis of the concept is to partition a predefined region into zones that consist of a combination of soil characteristics and deposition levels. The finding of chapter 4 that a metamodel may be used instead of the full model, in the calibration case, is used in each zone to test if a zone consists of similar types of lakes. Since the method does not use calibration it does not follow immediately that the usage of metamodels in a zone is allowed. This is clarified in the following:

The assumption is made that calibration is not needed if the model is able to make predictions of a <u>set of lakes</u> that are as accurate as the model would predict the quality of <u>one lake site</u>. Indeed, if a region consists of similar types of lakes, i.e be homogeneous, the deterministic[17] model result obtained for one single lake would be representative for the region; calibration would be obsolete. However a region does not consist of exactly similar kinds of lakes. Nor can zones be designed to fulfill exactly that condition. What can be achieved is to design zones that contain lakes that approximately exhibit a similar behavior in reaction to acid deposition. Therefore, the appropriateness of zones designed in that manner is tested by

[17]Note that for the application on one single lake many characteristics of the watershed can be represented by point estimates rather than by parameter ranges.

verifying if a metamodel may be applied instead of the model, i.e. test the fit of the metamodel predictions to the RAINS Lake model predictions.

The number of zones that result in the course of the analysis need not be fixed. Obviously, the method may result in zones within which model predictions have to apply to types of lakes that are similar under one kind of acid deposition conditions but become non-similar due to another deposition level. This is the kind of alternatives a policy maker usually may wish to evaluate. Such zones are subsequently partitioned again, until a good fit of the metamodel is obtained, and consequently appropriate policy predictions are provided. The method will be referred to in the following as the flexible zoning method.

In this chapter the flexible zoning method is described and illustrated.

5.2 FLEXIBLE ZONING

Method

The method of flexible zoning consists of 5 steps:

1. apply an uncertainty analysis using Monte Carlo Simulation on the model parameters when the model is applied to an arbitrary bounded region.
2. subdivide the ranges of the most important parameters to allow for different groups of soil characteristics, e.g. sensitive and insensitive zones, as follows. Four groups of parameters can be distinguished on the basis of their influence on the pH prediction, e.g parameters of which the increase leads to a pH increase or a pH decrease and parameters of which the decrease leads to a pH decrease respectively a pH increase. For example, in a parameter range of which the values lead to a pH decrease, a 'sensitive' range is taken close to the upper bound of the full value range whereas the 'insensitive' range will be chosen close to the lower bound. Combinations of subranges of different parameters that represent watershed characteristics in a region thus account for the 'sensitive' respectively

'insensitive' zones of watersheds in that region.

3. perform Monte Carlo Simulation on each of these zones subject to a few levels of the forcing function, i.e. acid deposition.
4. investigate the homogeneity (sampled watershed characteristics) of each zone by investigating the fit of a metamodel.
5. apply the cumulative distribution of model predictions to assess policy intentions, e.g. target loads, in a probabilistic way, i.e. assess the probability of lakes 'at risk' due to a target load.

The variability of predicted regional effects is obtained, as before, by applying PRISM (chapter 4) to the RAINS Lake Model. The region has been subdivided into 2 types of soil characteristics and 4 types of deposition levels. Thus an initial partition of 8 zones was obtained. An example is given to illustrate the method.

This example, worked out below, is based on one arbitrarily chosen region for which data is available at a similar level as shown in Table 4.1, and for which it was difficult to obtain a satisfactory calibration (see chapter 4), i.e. the Southern region of Norway. The most important parameters are however adapted using additional information from literature and other sources, as is explained below. Thus, the resulting parameter range or distribution become arbitrarily different from the ranges obtained originally for this region.

The starting point of the analysis is the performance of an uncertainty analysis, as explained in chapter 4, on region 1 of Norway exposed to depositions from the Official Energy Pathway scenario (chapter 2). The result is presented in Table 5.1.

Value ranges were sampled from all the ranges defined in region 1 of Norway except for the 4 parameters presented in Table 5.1. Extreme value ranges were imputed to soilt, calk, sibr and bsatb, with the aim of obtaining a combination of soil characteristics that were either sensitive (the lower range of soilt, sibr, and bsatb and the upper range of calk) or unsensitive (the opposite ranges of soilt, sibr, bsatb and calk) to sulfur deposition (see Table 5.2).

Table 5.1[1] Parameters explaining about 5% or more of SpH_{1980}[2]

Parameter Name3	Rank	%
SOILT	1	20.5
SIBR	2	15.6
BSATB	3	5.1
CALK	4	4.7

(Source: Hettelingh et al., 1988)
1Uncertainty analysis was performed on the RAINS Lake Module pH response in 1980, resulting from a combination of regional characteristics in South Norway and the Official Energy Pathways deposition pattern.
[2]As noted already, compared to Table 4.3 and 4.9 a shift of parameter uncertainties result when a combination of
regional characteristics and deposition pattern is changed.
[3]see Table 4.1 for an explanation of the parameters

These extreme value ranges have, for illustrative purposes, not be chosen such that their unification covers the entire range of the parameters. For example, in the region under consideration the soil thickness ranges from 0.05 to 5 meters. In the example only those watershed are investigated of which the soil thickness varies between 0.05 and 1 meter[18] for the sensitive and between 4 and 5 meters for the insensitive watersheds (see Table 5.2). The range of the silicate buffer rate (sibr) was derived from data sampled in the upper midwest lake area of the U.S.A by Schnoor et al. (1986). The base saturation is the fraction of the total cation exchange capacity available in soils (see equation 3.39 and 3.40). For the base saturation in the B-layer (bsatb) the upper half of the interval between 0 and 1 was assumed to be available in insensitive regions whereas a lower range was assigned in sensitive regions. The alkalinity constant (calk) is a function of the partial pressure of CO_2, the first acidity constant and of Henry's law constant (see also equation 3.11, 3.12 and 3.68).
The range of values of the alkalinity constant was defined (Cook, 1988) by applying information about these constants from the literature (Herczeg and Hesslein, 1984).

[18]The soil thickness is therefore 0.5 m. thicker than the A-layer defined in chapter 3.

Table 5.2 Parameter value ranges for sensitive and insensitive soil-characteristics

Parameter Name	Sensitive Minimum	Sensitive Maximum	Insensitive Minimum	Insensitive Maximum
soilt	0.05	1.0	4.0	5.0
sibr	0.04	0.067	0.067	0.64
bsatb	0.08	0.55	0.55	1.0
calk	6.21E-6	1.36E-5	4.98E-6	6.21E-6

(adapted from Hettelingh et al., 1988)

Yearly sulfur deposition scenarios of 0.0, 1.0, 5.0 and 10.0 $g/m^2/yr$ were simulated for both the above defined sensitive and insensitive regional types, covering the range of sulfur deposition widespread in Europe. Similar to section 4.4, the time horizon covered 1920 to 1980. Monte Carlo Simulations were performed on each of the 8 zones by sampling values from all ranges of the parameters, defined in Table 4.1, replacing the ranges of the parameters in Table 5.1 by the ranges of Table 5.2, and substituting the proper zone-deposition level. Then, an uncertainty analysis is performed on the Monte Carlo simulation results of the RAINS Lake Model in every zone. Finally, the most important parameters (explaining at least 1% of the variance of SpH_{1980}) are assessed to investigate the compatibility of the regional characteristics with the model results. In other words to test the homogeneity of the zone in which model results were obtained.

The statistical analysis performed on the behavior of the RAINS Lake Module in each of the 8 zones involves the application of metamodeling. The result in chapter 4.4 (equation 4.4) indicated that when calibrated to stable circumstances (soil characteristics and deposition pattern), a metamodel performs as well as the RAINS Lake model. Therefore, the fit of the metamodel to the sampled parameter values and model response obtained with PRISM, serves as an indication of the homogeneity of lake types in a zone. By lake type the combination is meant of areal parameters, for instance lake and catchment area.

Two metamodels were fitted to the Monte Carlo Simulation results:

$$SpH_{1980} = a_0 + a_1*P_1 + \ldots + a_n*P_n \qquad (5.1)$$

$$SpH_{1980} = b_0 + (b_1*P_1 + \ldots + b_m*Pm) + (c_1*(P_1)^2 + \ldots + c_m*(P_m)^2) + (d_1*P_1*P_2 + \ldots + d_M*P_{m-1}*P_m) \quad (5.2)$$

where,

SpH_{1980} = simulated pH in 1980 using the RAINS Lake module in a zone

P_i = most important parameter i, i=1,...,m,
max(m)= 19, (Table 4.1), min(m) = 5 (most important parameters).
a,b,c,d = coefficients

Equation 5.1 describes the Monte Carlo simulation pH response of the RAINS Lake Model as linear function of the most important parameters in every zone. Equation 5.2 includes quadratic and cross term elements of the top 5 most important parameters in every zone.

The importance of parameters was evaluated using the concept of partial R^2 (see chapter 2; equation 2.16). The fit of equation 5.1 was evaluated by means of the multiple correlation coefficient. For the fit of equation 5.2 an adjusted multiple correlation coefficient was defined as follows:

$$AR^2q = (TSS - ESSq)/TSS \quad (5.3)$$

where,

AR^2q = Adjusted multiple correlation coefficient of eq. 5.2

TSS = Sum of squares about the mean (Total Sum of Squares) of equation 5.1

ESSq = Sum of squares due to regression (Error Sum of Squares) of equation 5.2

Results of an application of flexible regionalization

An uncertainty analysis on the sensitive and insensitive zones resulted in a ranking of parameters that are respectively shown in Figure 5.1 and Figure 5.2.

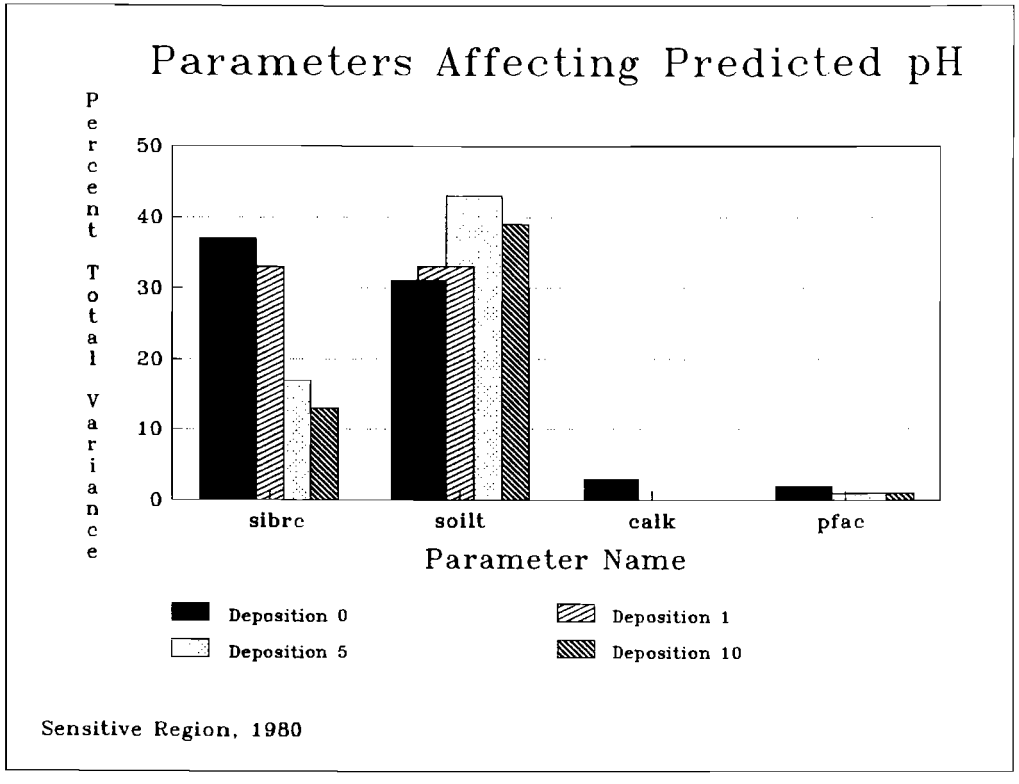

Figure 5.1 Parameters most affecting predicted pH in the sensitive subregion.

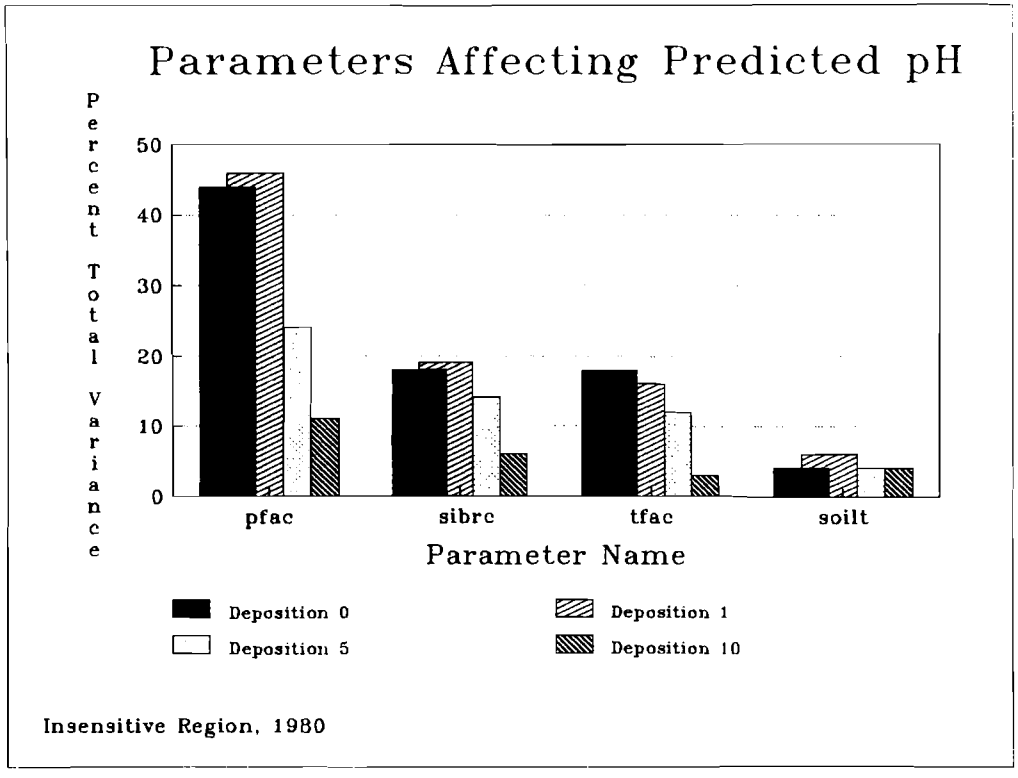

Figure 5.2 Parameters most affecting predicted pH in the insensitive subregion.

The conclusions of chapter 4 are clearly confirmed: a shift in uncertainties occurs whenever deposition patterns and regional characteristics vary. In the insensitive subregion the first most important parameter is pfac[19] explaining most of the pH variance in the 1 deposition zone (46%) and the least in the 10 deposition zone (11%). In the sensitive subregion, however, soilt is the most important parameter in resp. the 1 (44%), 5 (43%) and 10 (39%) deposition zone, but is replaced by sibr in the 0 deposition zone (partial R^2 of sibr is 37% and of soilt 31%). The conclusion is that within a fixed region different zones can be distinguished leading to varying model parameter uncertainties.

Table 5.3 shows mean, median, minimum and maximum values of predicted pH (SpH_{1980}) in every zone. An overview of the statistics and a display of the cumulative distribution of SpH_{1980} for all 8 zones is given in appendix III.

Table 5.3 Mean, Median, Minimum and Maximum SpH_{1980} by zone

Region Type	Yearly Deposition Levels ($g/m^2/yr$) 0	1	5	10
Insensitive				
Mean	7.63	7.47	7.37	7.18
Median	7.61	7.46	7.38	7.31
Minimum	7.17	6.90	4.69	3.97
Maximum	8.07	7.93	8.01	7.93
Sensitive				
Mean	7.02	5.82	4.72	4.48
Median	7.20	6.10	4.05	3.97
Minimum	4.46	4.0	3.82	3.58
Maximum	8.07	7.91	7.82	7.53

(adapted from Hettelingh et al., 1988)
[1] SpH1980 is the lake pH in 1980 predicted by the Monte Carlo simulation of the RAINS Lake Module. The statistics shown are based on 500 Monte Carlo simulations.

As expected, for a given deposition level, predicted pH values tend to be lower in the sensitive subregion. Furthermore, predicted median pH values in the sensitive subregion decrease markedly with increasing deposition (7.2 under 0 $g/m^2/yr$ deposition to 3.97 under 10 $g/m^2/yr$),

[19] See table 4.1 for the meaning of parameter abbreviations.

whereas predicted median pH values in the insensitive region remain relatively constant (> 7.31).

Examination of predicted pH ranges (Table 5.3) show that the ranges of pH values are relatively constant in the sensitive region for all deposition levels, ranging from 4.46 to 8.07 under zero deposition and from 3.58 to 7.53 under a deposition scenario of 10 $g/m^2/yr$ (see also appendix III). Predicted pH in the insensitive region ranged from 7.17 to 8.07 under zero deposition, and from 3.97 to 7.93 under deposition of 10 $g/m^2/y$. The relatively stable range of pH values in the sensitive region suggests that this region responds in a homogeneous fashion to sulfur deposition (i.e., the effect of random variation among watersheds is relatively small), while the insensitive region at high deposition levels displays a heterogeneous response.

Another indication of the relative heterogeneity of the insensitive region is provided by the multiple correlation coefficients of the response surface models. Table 5.4 shows the magnitudes of the multiple correlation coefficients for the two metamodels for each zone.

Table 5.4 Multiple Correlation Coefficients for the Standard (R^2) and for the Quadratic (AR^2q) metamodel for every zone.

Region Type	Yearly Deposition Levels ($g/m^2/y$) 0	1	5	10
Insensitive				
R^2	0.90	0.90	0.67	0.30
AR^2q	0.83	0.85	0.58	0.30
Sensitive				
R^2	0.79	0.82	0.67	0.57
AR^2q	0.86	0.83	0.86	0.77

(adapted from Hettelingh et al., 1988)

The percentage of the variance explained by the metamodels decreases substantially in the insensitive subregion with increasing sulfur deposition. Multiple correlation coefficients for the quadratic and standard models decrease from 0.90 and 0.83 under zero deposition to 0.30 under the 10 $g/m^2/y$ scenario. In contrast, the magnitudes of the multiple correlation coefficients for the sensitive subregion show a much smaller decrease with increasing deposition levels. Even under the

10 g/m^2/y deposition level, the two metamodels explain at least 50% of the variability in predicted pH. Thus, as the level of sulfur deposition increases the predicted pH in the insensitive region shows an increased range of variability in predicted lake pH.

The relatively low multiple correlation coefficient for the insensitive subregion under 5 and 10 g/m^2/yr deposition scenarios can be attributed to heterogeneity within the regional type. When a critical pH level is chosen at which damage to watershed flora and fauna starts, i.e. 6.5 (see Figure 1.2), then two subregions (Table 5.5) can be distinguished within the insensitive-10 g/m^2/yr zone.

Table 5.5 Physical Characteristics of Subsets of the Monte Carlo Simulation for the insensitive-deposition of 10 g/m2/yr zone

Parameter Name	pH < 6.5			pH ≥ 6.5		
	min	max	mean	min	max	mean
LAKAR	11.3	604.7	108.9	1.1	937.0	107.1
RATCL	1.0	11.6	3.9	1.1	747.1	46.1
SpH1980	3.97	6.49	5.27	6.51	7.93	7.32

(adapted from Hettelingh et al., 1988)

Comparison of the sets of parameter values of these two subregions shows that the pH < 6.5 subregion comprises small catchment lakes (mean clrat of 3.9), whereas the pH ≥ 6.5 subregion comprises large catchment lakes (mean clrat of 46.1). Application of equation 5.1 to each of these subregions results in correlation coefficients of 0.77 for the pH<6.5 subregion and 0.69 for the pH ≥ 6.5 subregion.

5.3 POLICY MAKING WITH FLEXIBLE ZONING

As mentioned in chapter 1, threshold levels for pollution effects are becoming increasingly important for decision making, but the analysis of uncertainties shows that it is difficult to apply concepts of thresholds within heterogeneous regions (i.e, when regional boundaries are arbitrarily established). Two kinds of thresholds have

been distinguished (see section 1.2): (1) target loads which are the policy related thresholds for deposition and acidification levels within a country or region; and (2) critical loads which are deposition levels which produce a predetermined level of effects. From the foregoing it is clear that the establishment of a deterministic threshold for a predefined region may lead to wrong policy decisions, because of the varying ways in which watersheds are influenced by deposition patterns and regional characteristics. Calibration, as demonstrated in chapter 4, may not alleviate this fact.

Flexible zoning in combination with a probabilistic evaluation of watershed quality, may improve such policy assessments. This is illustrated by inspection of the cumulative model response distributions (appendix III) in every zone.

When for example a target load is assumed of respectively 0, 1, 5 and 10 $g/m^2/yr$, and when, as above, a threshold pH of 6.5 is taken as indicator for water quality, the assessment of the cumulative probability functions (appendix III) for each of the 8 zones provides a policy maker with an estimation of sets of lakes under risk within a zone (see Table 5.6).

Table 5.6 Assessing target loads to 8 lake zones within a zone at a critical pH indicator of 6.5

Region Type	Yearly Deposition Levels ($g/m^2/y$) 0	1	5	10
Insensitive				
% of lakes with pH<6.5	0	0	3	5
Sensitive				
% of lakes with pH<6.5	20	55	85	87

The resolution of Table 5.6 can easily be improved by incorporating more deposition levels in the analysis and by investigating the percentiles from appendix III at different other pH levels that might be considered relevant from a policy makers point of view.

5.4 CONCLUDING REMARKS

This chapter has evaluated the potential for policy making of replacing an arbitrarily bounded region by different zones for the assessment of environmental quality, i.e. watershed quality. It has been shown that heterogeneity within a region should be considered when developing policy guidelines concerning the effect of target deposition levels. An example region was disaggregated to 8 zones and Monte Carlo simulation of the RAINS Lake Module results was applied.

The method of flexible regionalization consists of the following steps:

1. perform an uncertainty analysis on model parameters when applied to the arbitrarily bounded region,
2. subdivide the ranges of the most important parameters (as determined in step 1) into ranges which can be recombined into zones that are either sensitive or insensitive to acid deposition,
3. perform Monte Carlo simulation for each of these zones under varying deposition levels. Four deposition levels were chosen for this study of 0, 1, 5 and 10 $g/m^2/yr$,
4. investigate the homogeneity of each of the zones, and determine if the homogeneity criterion is satisfied -- if not, then further subdivisions may be necessary. Two metamodels were fit to the simulation results of the RAINS Lake Module and the goodness of fit was used as indicator of homogeneity within each zone, and
5. assess different target (or critical) loads on the cumulative distribution of the response in every zone, leading to a probabilistic evaluation of policy intentions.

The advantage of the method is that it focusses on predictions of sub-systems within a region for which data samples are not sufficient to identify potential environmental damage. Monitoring programs executed at a point in time on an arbitrarily defined region, may not lead to the desired insight concerning environmental quality. This is

especially true if the most important forcing function of the system (deposition) has a spatial or temporal gradient. Calibration procedures may not prove sufficient (see chapter 4) to circumvent the drawbacks of policy assessments to environmental systems (watersheds) that exhibit heterogeneous characteristics within a predefined region.

Validation (against data) of model results of flexible zones is only possible if measurements of model response are available for every zone. In the absence of these measurements, the method of flexible zoning provides a relative distinction of sets of environmental systems (watersheds) being at risk, i.e. compared to model response in other zones.

Model predictions thus become less dependent on the calibration and aggregation errors that may result (see chapter 4) from the choice of a fixed zoning system.

6. APPLICATION TO DUTCH WATERSHEDS

6.1 INTRODUCTION

Over the last decade the quality of the natural environment in the Netherlands has become subject to increasing concern. An overview of the causes, the effects and the projected levels of its pollution has recently been published (RIVM, 1988) in view of the preparation of 'the National Environmental Policy Plan' (NMP). Acidification, of which an important share in the Netherlands is caused by an over-production of livestock manure, is one of the important environmental policy issues. Deposition of ammonium-N, nitrate-N and sulfate-S has led to a decrease of the vitality and diversity of species in heath, forests, small lakes and moorland pools (RIVM, 1988, pp. 112). The occurrence of particular plant species in small surface waters at different pH levels has shown that acidification has increasingly affected the quality of Dutch surface waters after 1950 (Roelofs and Schuurkes, 1983; Roelofs et al., 1984).

In the period 1983-1984 the quality of sensitive (< 100 ha and rather shallow) surface waters has been monitored
(see also Schuurkes and Leuven 1986, Schuurkes 1987 and Leuven 1988). The results (Kersten, 1985) of these field measurements show that about 90% of the lakes have a pH lower than 7, and 50% of the investigated surface waters have a pH lower than 4.2 (Schuurkes and Leuven, 1986, pp. 16). A percentage between 4.6 and 10.2% of the total Dutch clear water surfaces, excluding the Ysselmeer, have been acidified (Schuurkes and Leuven, 1986, pp. 27).

On the basis of dose-effect relationships, Schuurkes (1987) found that the threshold at which no apparent change in the water quality occurs lies at an acidity level of 250 mol/ha/yr. Leuven (1988) estimates that the critical load lies between 250 and 800 mol/ha/yr for waters having low calcium levels.

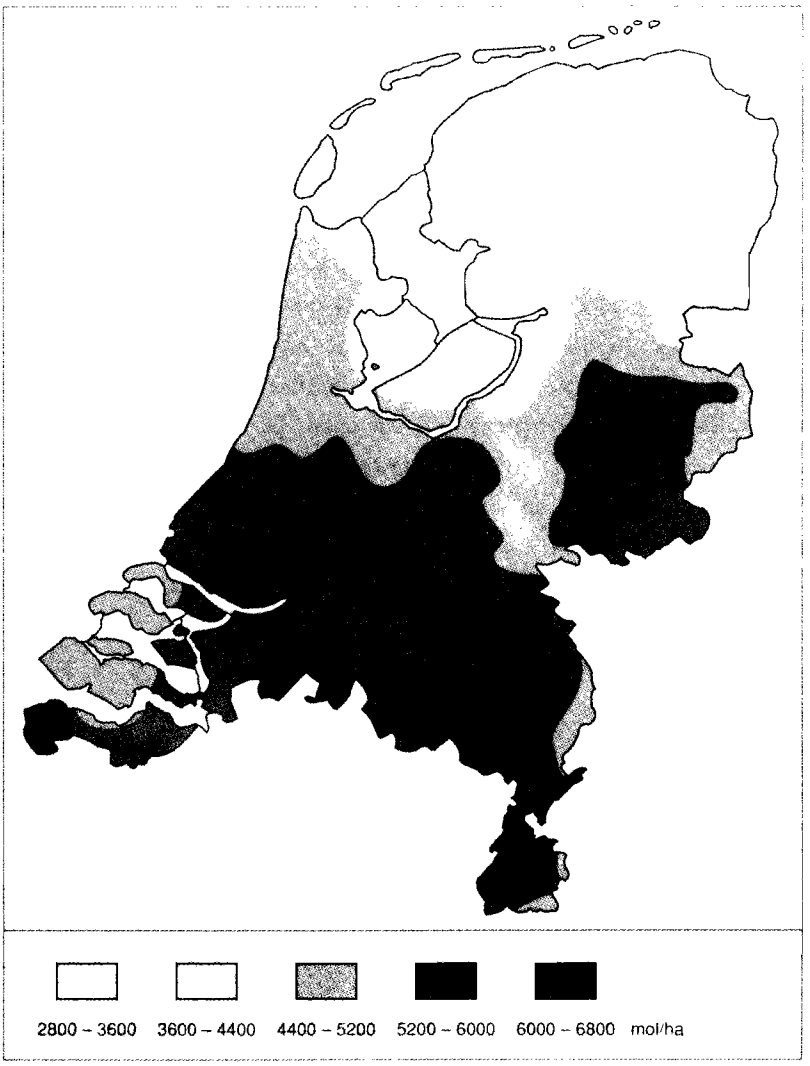

Figure 6.1 Deposition of potentially acidifying constituents in the Netherlands in 1986 in mol/ha (Source: RIVM, 1988, pp. 109).

In sandy soils a critical load that lies in the range of 500 to 700 mol/ha/yr was estimated by the RIVM (1988, pp.103), and a level below 600 mol/ha/yr was proposed by de Vries (1988).

Note that the assumed critical load for soils is similar to the one assumed for the maintenance of a healthy water quality. However, inspection of the deposition of potential acid in 1986 (Figure 6.1) shows that these levels have been exceeded, especially in the south-east and east of the Netherlands where most of the acidified small lakes and sensitive soils are found.

Simulation models have been applied to describe soil acidification in the Netherlands. ILWAS was modified and calibrated to an acid sandy forest soil (van Grinsven, 1988) and the REgional Soil Acidification Model (RESAM, see de Vries, 1987) was applied to COROP regions. However, regional simulation to estimate and describe lake acidification in the Netherlands has not been performed yet.

This chapter will provide a first attempt in this field, by the use of the RAINS Lake Model to describe the pH levels on a regional scale in the Netherlands. It will be shown that the method of flexible zoning, as proposed in chapter 5, offers insight into acidification of broad watershed areas in the Netherlands when deposition levels are varied within the ranges currently prevailing over the Netherlands.

A description of the data is given, then the adaptation of the RAINS Lake model is discussed and agreement with the Dutch data, particularly the influence of Nitrates on the acidification process, is tested. Finally the results are presented, and the simulated effect of a range of critical loads is determined.

6.2 DUTCH WATERSHED DATA

Dutch data on soil and lake characteristics had to be collected to serve as input to a RAINS Lake Model application in the Netherlands. Lake characteristics were provided by the Laboratory for Aquatic Ecology (Maessen, 1988) of the University of Nijmegen. Data on geochemical soil characteristics were obtained from de Vries (1989) who

extracted the information from the Information System on soils of the Netherlands Soil Survey Institute (STIBOKA).

Table 6.1 RAINS Lake Model parameter statistics and ranges for Dutch surface waters and soil characteristics[1]

Code name[2]	Mean	Stand.Dev.	Min.	Max	Unit
lakar	2.0	23.07	0.1	80	ha
clrat	1.5	0.57	0.0001	2.0	ratio
ldept	1.0	0.29	0.01	2.0	m
soilt	1.0	0.79	0.25	3.0	m
slope	0.0005	2.89e-04	0.0	0.001	m/m
sibr	0.02	0.01	0.01	0.03	eq/m^3 yr
cecA[3]	54.6	40.3	2.6	356.6	eq/m3 yr
bvA[4]	0.10	0.046	0.034	0.216	fraction
fcap	0.15	0.13	0.05	0.50	fraction
bvC[4]	0.065	0.039	0.018	0.206	fraction
pCO_2	0.02	0.01	0.01	0.04	atm
foco[5]	0.3	3.33e-03	0.2	0.4	fraction
sulre[6]	0.4	0.12	0.2	0.6	m/yr
fofi	2.25	0.43	1.5	3.0	factor
basca[7]	0.65	0.09	0.5	0.8	fraction
tfac	0.5	0.29	0.	1.0	fraction
pfac	0.5	0.29	0.	1.0	fraction
cecB[3]	35.0	35.0	0.0	249.2	eq/m^3 yr
cecC[3]	6.52	10.87	0.0	142.46	eq/m^3 yr
bvB[5]	0.065	0.039	0.018	0.206	fraction
cond	0.2	0.12	0.1	0.5	m/yr
NHeq[8]	0.10	0.020	0.0579	0.1429	eq/m^2 yr
NOeq[8]	0.09	0.02	0.06	0.12	eq/m^2 yr
fnit[8]	0.875	0.07	0.75	1.0	fraction
nitup[8]	0.07	0.01	0.057	0.086	eq/m^2 yr

[1] Data were collected from various unpublished sources (see text) and reflect the author's best understanding of the Dutch watershed systems.
2See Table 4.1 for a description of most of the code names. Other code names are explained in this chapter.
[3] The cation exchange capacity in the upper 0.15 m of the soil.
cecB applies to the layer between 0.15 and 0.5 m and cecC is the cation exchange capacity in layers between 0.5 and 2.5 m.
[4] Basesaturation in the upper 0.15 m of the soil. Similarly bvB applies to 0.15-0.5 m whereas bvC applies to 0.5-2.5 m.
[5] Forest coverage
[6] In-lake sulfate retention
[7] Correction for the Base-cation deposition of SO_4 loading
[8] See next section about inclusion of Nitrate-N and Ammonia-N in the RAINS Lake model.

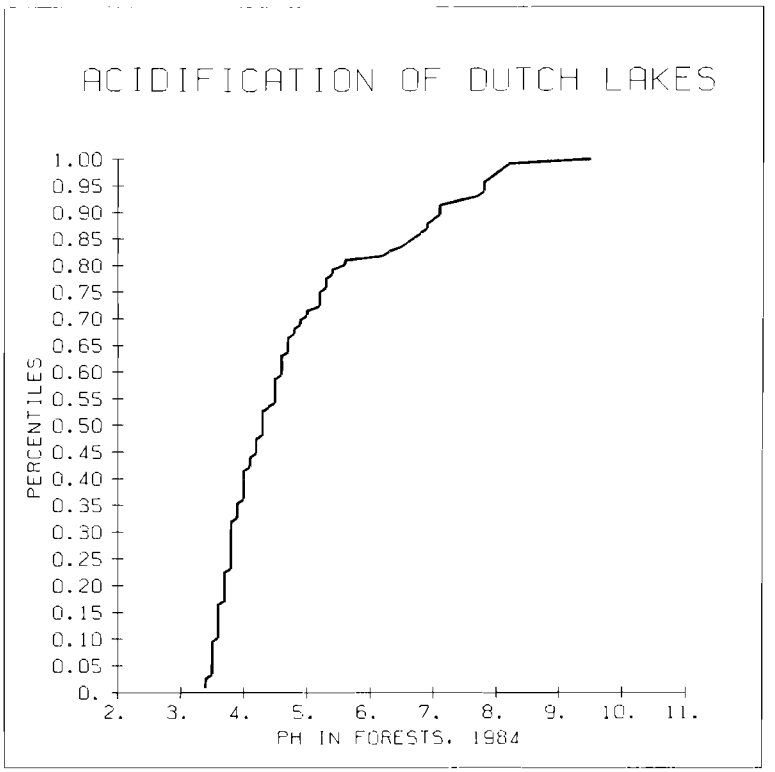

Figure 6.2 Cumulative distribution of measured pH in forested Dutch areas sampled on 1984 (digitized from Kersten, 1985).

Climatologic data on precipitation and temperature were obtained from MÜller (1982, pp. 20). The measurements of the Dutch weather stations, i.e., Eelde, Den Helder and De Bilt, were used in order to obtain precipitation and temperature ranges representative for the Netherlands.

The Monte Carlo input parameter ranges that were thus compiled are presented in Table 6.1. Table 6.1 illustrates that Dutch catchments are rather small (maximally twice a lake area), that the lakes are shallow (maximally 2 meters) and that the Dutch soils are thin (maximally 3 meters).

Kersten (1985) sampled the physical and chemical characteristics of about 164 Dutch watersheds in 1984. A majority of the sampled watersheds, i.e., about 116, are situated in forested areas. The cumulative distribution of the pH for these catchments has been digitized from Kersten (1985) and the result is displayed in Figure 6.2. Since damage to the biota within watersheds is assumed to be attained at a pH level below 5 (see chapter 1, Figure 1.2), it can be seen from Figure 6.2 that about 70% of Dutch watersheds in forested areas are at risk.

Dutch lakes are different from the lakes in Scandinavian countries from a geo-physical as well as from an acidification point of view. Soil processes may have a limited impact on Dutch lake acidification and the causes of acidification do not originate mainly from sulfur deposition. The different geo-physical conditions are incorporated in the Monte Carlo parameter ranges for the RAINS Lake model, but the influence of Nitrogen is not part of the RAINS Lake Model.

The next section will discuss the changes made to the RAINS Lake Model in order to take the special Dutch conditions into account.

Adaptations to the RAINS Lake Model

Changes to the RAINS Lake Model were made to (1) incorporate 3 soil-layers instead of 2 and (2) to take N-deposition into account.

STIBOKA (de Vries, 1988) distinguishes three soil-layers: the A-layer (0-0.15 m), the B-layer (0.15-0.5 m) and the C-layer (deeper than 0.5) each with unique cation exchange capacity (CEC) and base-saturation characteristics (see also chapter 3). Thus, the A-layer in

RAINS is equivalent to the A and B layers defined by STIBOKA, whereas the B-layer in RAINS is equivalent to the C-layer defined by STIBOKA. The threefold partitioning of the soil has consequences for the equations 3.37, 3.38, 3.39 and 3.40 of RAINS described in chapter 3. If for example a soil thickness (soilt) is greater than 0.5 meter, the RAINS equations become respectively:

$$A^{cec} = 0.15*cecA + 0.35*cecB \quad (6.1)$$

$$B^{cec} = (soilt-0.5)*cecC \quad (6.2)$$

$$A^{eat} = 0.15*cecA*bvA + 0.35*cecB*bvB \quad (6.3)$$

$$B^{eat} = (soilt-0.5)*cecC*bvC \quad (6.4)$$

where, the parameters have values from the ranges specified in Table 6.1.

The meteorologic part of the RAINS model was extended and tested to incorporate N-deposition with the following modification of equation 3.17 (de Vries 1989):

$$(D^{load})_t = (D^{tot})_t - (D^{bc})_t + NOeq + (2*fnit-1) * NHeq + - nitup \quad (6.5)$$

The purpose of equation 6.5 is that the extent to which nitrification has occurred (fnit varies between 0.75 and 1, see Table 6.1), will affect the ammonia level that contributes to acid stress. In addition it is assumed that due to organic processes nitrogen uptake can vary between 10 and 11 kg/ha[20]. A detailed description of a soil acidification model that incorporates these processes can be found in de Vries et al. (1989).

Equation 6.5 was compared to the unmodified version (equation 3.17) by running the RAINS LAKE model using all the input-ranges of Table 6.1. Note, that equation 6.5 becomes equal to equation 3.17 when 0 is substituted for NOeq, NHeq and nitup. The model was started in 1900 using historical European emissions (Fjeld, 1976 quoted in Whelpdale, 1987) that were dissaggregated to country level contributions in 1980 (see also chapter 4).

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[20] 10 kg/ha N uptake corresponds to 1 g/m^2 = 0.07 eq/m^2 (atomic weight of N is 14).

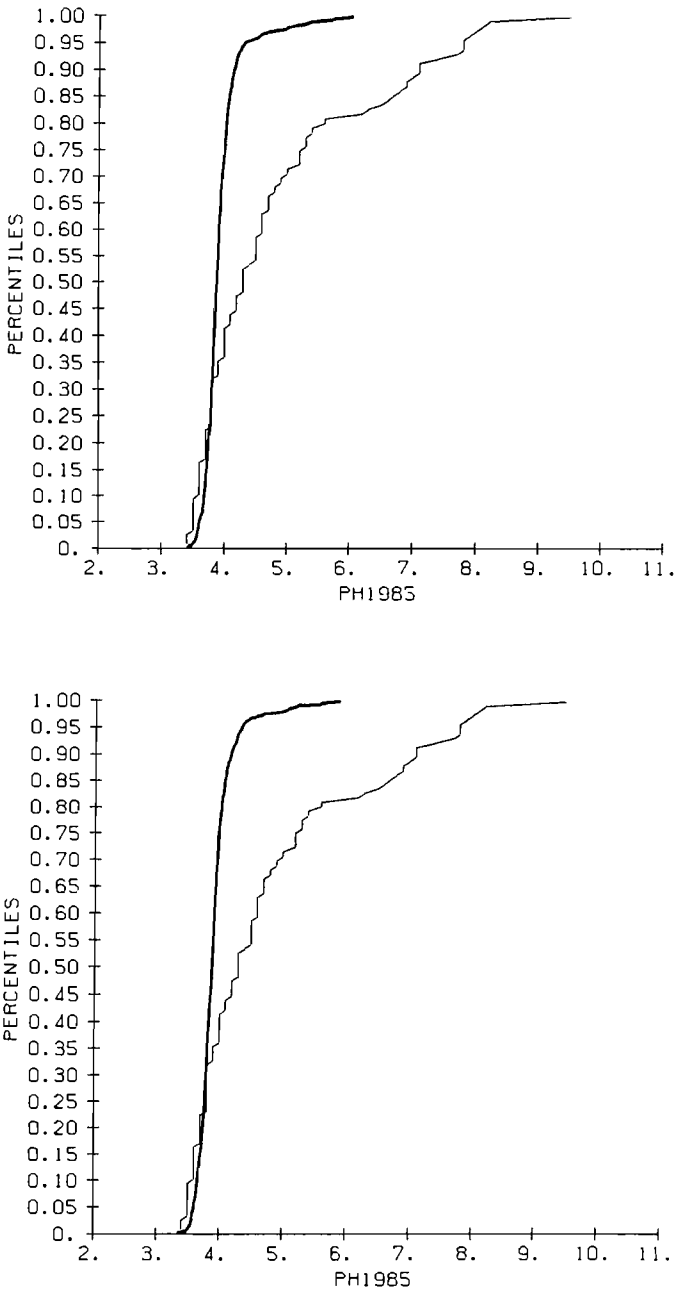

Figure 6.3 Simulated pH (thick graphs) in 1985 excluding N (a) and including N (b), compared to the cumulative distribution of measurements.

The range of NO_x and NH_3 depositions over the Dutch area were taken from the RAINS model in 1985 and were assumed to be unchanged since 1900. The result of the lake acidification in the Netherlands in 1985 due to equation 3.17 is expressed in Figure 6.3a, and the acidification due to equation 6.5 is shown in Figure 6.3b. It is clear from comparing Figure 6.3a to Figure 6.3b that equation 6.5 does not affect the RAINS Lake Model predictions of lake acidification in the Netherlands. Therefore, instead of equation 6.5, total potential acid stress (which is due to N and S deposition) may be used as input to model equation 3.17. Total potential acid stress is implemented in step 2 of the investigation method, applied to the Dutch lakes, which is described in the next section.

6.3 REGIONAL WATERSHED ACIDIFICATION IN THE NETHERLANDS

Introduction

This section discusses the RAINS Lake Model predictions of pH, for zones in the Netherlands that consist of a combination of deposition ranges and of sensitive and insensitive catchment characteristics. In chapter 5 an example was given of the method of flexible zoning in contrast to the results described in chapter 4 that were based on calibration of the model in predefined regions. For the Netherlands predefined regions have not been defined yet for managing large scale surface water quality. An application of the method of flexible zoning will therefore be used to identify the kinds of catchment zones for which water quality management is particularly useful, according to RAINS Lake Model predictions.

The method

Recall from chapter 5 that the method of flexible zoning consists of 5 steps that are interpreted as follows for the Dutch application:

1. Performing an uncertainty analysis of the model and the Dutch data.

2. Subdividing the value ranges of the most important parameters into watershed zones that increase the resolution of the model predictions, i.e. recognize kinds of watersheds that are identified by the model to be at risk, relative to different levels of potential acid deposition. Four zones of acid deposition have been defined in accordance with the deposition pattern displayed in Figure 6.1, i.e. 3600-4400, 4400-5200, 5200-6000 and 6000-6800 mol/ha.
3. Performing Monte Carlo Simulations with the model for every zone of watershed characteristics in combination with a range of potential acid deposition.
4. Investigating the R^2 of pH predictions when regressed on the most important parameters in each zone. Decide if a further breakdown of zones is necessary (if so, repeat steps 2-4).
5. Compare the model predictions in every zone to the response of the model in the same zone when a critical load range of 250-800 mol/ha/yr (see section 6.1) is specified for model simulations.

The modified RAINS Lake Model is then run from 1900 to 1985 and from 1900 to 2040. The first period is used to investigate model predictions in 1985 when each of the four deposition ranges remain unchanged since 1900. In addition, predictions of the pH in 1985 are also obtained for the artificial case that critical loads have been deposited on the Dutch surface since 1900. The latter pH predictions in 1985 will be referred to as Critical Load Acidification. The Critical Load Acidification level can be compared to future acidification levels that result from policy measures to abate emissions. An example of interesting policy measures are those that lead to deposition levels being equal to critical loads. If policy measures lead to a deposition of critical loads from 1990 and future years, the question is addressed how long it will take before the acidification 'recovers' to the Critical Load Acidification. The comparison of the Critical Load Acidification with other model predictions thus provides a relative measure of interpreting the effect of policy measures with the RAINS Lake Model.

Results

Results of an uncertainty analysis of the RAINS Lake Model pH prediction in 1985 including N (Figure 6.3a) and excluding N (Figure 6.3b) show that the forest filtering factor and sulfate retention parameter are the two most important parameters. Soil thickness plays a minor role in the Netherlands in comparison to its importance in other countries (see chapter 4 and 5). Table 6.2 summarizes the ranking of the model parameters that have a partial R^2 (see chapter 2) of at least 1%.

In the analysis including N as well as in the analysis excluding N, an R^2 of about 0.4 was obtained when regressing the predicted pH in 1985 against all the parameters listed in Table 6.1 and yearly sulfur deposition. This low R^2 indicates a poor fit between the simulated pH and the sampled values of the soil and catchment characteristics, of the climatology and of the deposition.

Figure 6.3a shows that the model proves to be capable of predicting lake effects below the 0.35 percentile of the measurements. About 95% of the simulated pH lies in a range of 3.4 to 4.4. In 5% of the simulations a pH between 3.4 and 5.9 is simulated.

To avoid attaching too much absolute value to these predictions, a comparison was made with the Critical Load Acidification in 1985 (Figure 6.4).

Table 6.2 Parameters explaining more than 1% of the variance of the predicted pH in 1985 in Dutch watersheds.

Name	including N (%)	excluding N (%)
fofi	11	7
sulre	8	8
soilt	5	5
basca	3	-
tfac	3	3
sibr	3	-
foco	2	2
clrat	2	1
pfac	-	3

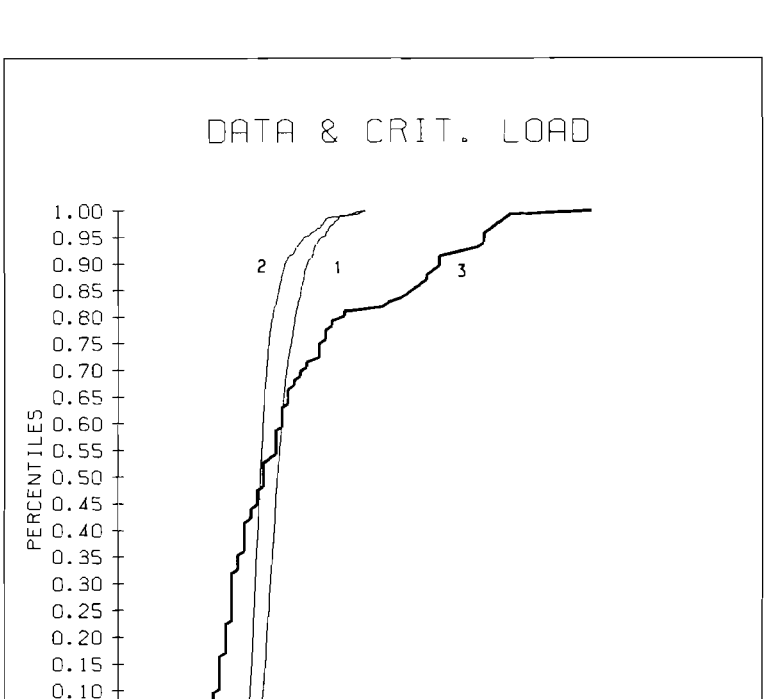

Figure 6.4 Cumulative distributions of the simulated pH in 1985 due to the minimal (graph 1) and maximal (graph 2) Critical Load Acidification compared to the cumulative distribution of the measurements (3).

The pH range simulated when Dutch catchment characteristics are used for model inputs in combination with minimal and maximal critical loads since 1900, covers a pH of 3.9 to 5.9.

It is obvious from Figures 6.3a, 6.3b and 6.4 that the RAINS Lake Model is unable to predict the 20% of the measured pH values that lie between 5.9 and 9.5, given the current ranges within which Dutch catchment and soil characteristics have been monitored. Even the input of a critical load range (Figure 6.4) did not result in a prediction of these high pH values. The choice of deposition levels below the critical loads would not be supported by current understanding about the thresholds of deposition above which damage may occur.

The next step (step 2) is therefore to subdivide the Dutch area into four ranges of total acid deposition. In addition the catchment properties parameters are partitioned, around the mean[21], into a sensitive and an insensitive range if a parameter value respectively leads to a decrease or increase[22] of the simulated pH. The parameters ranges to be partitioned are not only chosen on the basis of their ranking in Table 6.2, as consequence of the low R^2 mentioned before. For example the weathering rate, subject to other catchment and deposition characteristics, has been an important parameter in past analyses (chapter 4 and chapter 5) and is therefore included as well. In addition the small Dutch catchments give rise to the assumption that, no matter how the zoning is done, soil buffering will have little effect on Dutch watershed acidification. This assumption is tested by including the Cation Exchange Capacity of both the Dutch A and B-layer as a zoning criterium. The result of the partitioning of the parameters is shown in Table 6.3.

An uncertainty analysis of the RAINS Lake Model in each of the zones resulted in a ranking of important parameters that is displayed in Table 6.4.

Table 6.4 displays how uncertainties differ in both the sensitive and insensitive zone. In the sensitive zone the temperature factor

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[21] It seems reasonable to use the average of sampled values for the distinction of two classes,i.e below the mean and above. Other statistics need not be excluded but make less sense.

[22] This is done by investigating the sign of the regression coefficients.

Table 6.3 Definition of sensitive and insensitive catchment characteristics.

Parameter name	Sensitive min.	Sensitive max.	Insensitive min.	Insensitive max.
fofi	1.0	1.5	1.5	3.0
sulre	0.2	0.4	0.4	0.6
soilt	0.25	1.0	1.0	3.0
basca	0.65	0.8	0.5	0.65
tfac	0.0	0.5	0.5	0.1
sibr	0.01	0.02	0.02	0.03
foco	0.2	0.3	0.3	0.4
pfac	0.5	1.0	0.0	0.5
ceca	2.6	54.6	54.6	356.2
cecb	0.0	35.0	35.0	249.2

Table 6.4 Uncertainty analysis in Sensitive (I) and insensitive (II) zones

Zone	Name	Potential acid deposition (mol/ha) 3600-4400	4400-5200	5200-6000	6000-6800
I	clrat	0.28	0.31	0.34	0.36
	tfac	0.31	0.30	0.29	0.29
	basca	0.12	0.12	0.12	0.12
	sulre	0.08	0.08	0.08	0.08
	pfac	0.06	0.06	0.06	0.05
	R^2	0.92	0.95	0.96	0.97
II	tfac	0.36	0.35	0.35	0.34
	fofi	0.15	0.17	0.18	0.19
	soilt	0.14	0.10	0.07	0.06
	pfac	0.06	0.06	0.06	0.06
	R^2	0.81	0.79	0.77	0.75

(tfac) and the catchment to lake ratio (clrat) explain from 59% to 65% of the variance of simulated pH of 1985 in, respectively, the lowest and highest potential acidic deposition ranges. In the insensitive zone, however, the forest filtering factor (fofi) replaces the importance of the catchment to lake ratio (clrat). In this zone tfac and fofi account for 51% and 53% in respectively the lowest and highest deposition range.

The multiple correlation coefficient (R^2) indicates that a response surface relationship containing the most important parameters fits the simulated pH in 1985 better than without zoning (Table 6.2). In other words, the Monte Carlo Simulations lead to a cluster of predictions that can be approximated by a linear function. Therefore flexible zoning has allowed the simulation of types of catchments with comparable characteristics. An analysis of the sampled catchment characteristics did not exhibit a criterion by which a zone could be further disaggregated, as was demonstrated in the example described in chapter 5.

The prediction of the pH in 1985 in the sensitive and insensitive zones are presented in Figures 6.5 and 6.6. In each figure the cumulative distributions of the Critical Load Acidification (graph 1) of the measurements (graph 2), and the predicted pH in the lowest (graph 3) and highest (graph 4) potential acid deposition zones is plotted. The two middle deposition zones (4400-5200 and 5200-6000) are not shown, but lead to pH distributions that lie between graph 3 and graph 4. Note that in Figure 6.6 the ph predictions in the 3600-4400 zone show a close resemblance with the data below the 70 percentile.

Data were not available to this study to check the kind of catchments that were actually measured. However, the differences between the simulated pH in the sensitive (Figure 6.5) and insensitive (Figure 6.6) zones support the statement made earlier that the resolution of the model predictions is increased by partitioning the region. Policy decisions evaluated by model predictions from the flexible zones can be linked to possible responses of catchment systems over the Netherlands. This assertion was tested by assuming that measures are taken throughout Europe leading to a deposition that lies

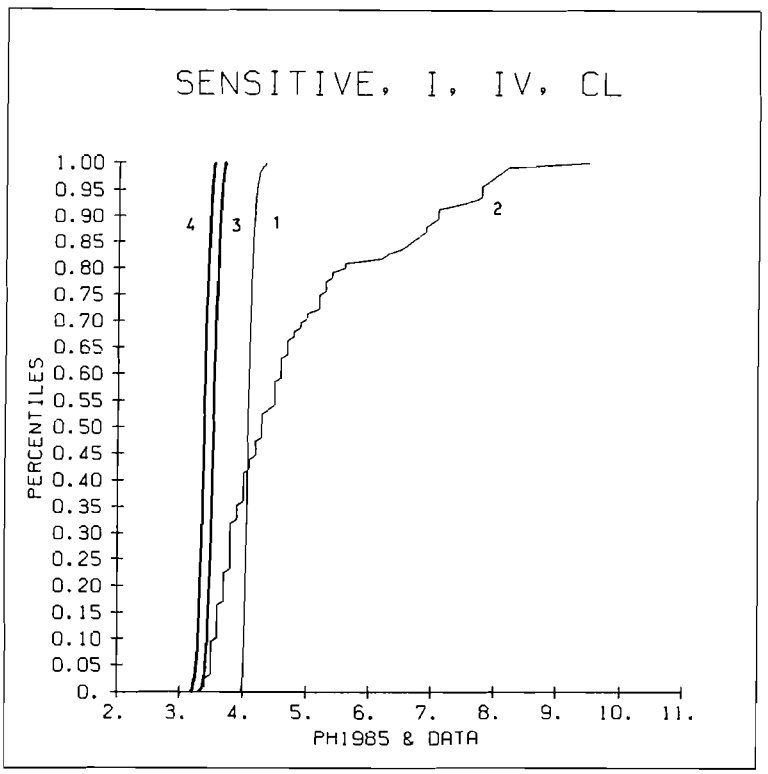

Figure 6.5 Cumulative distributions of the simulated pH in the sensitive class in 1985 due to the Critical Load Acidification (graph 1), to the lowest deposition range (graph 3) , to the highest deposition range (graph 4), compared to the cumulative distributions of the measurements (graph 2).

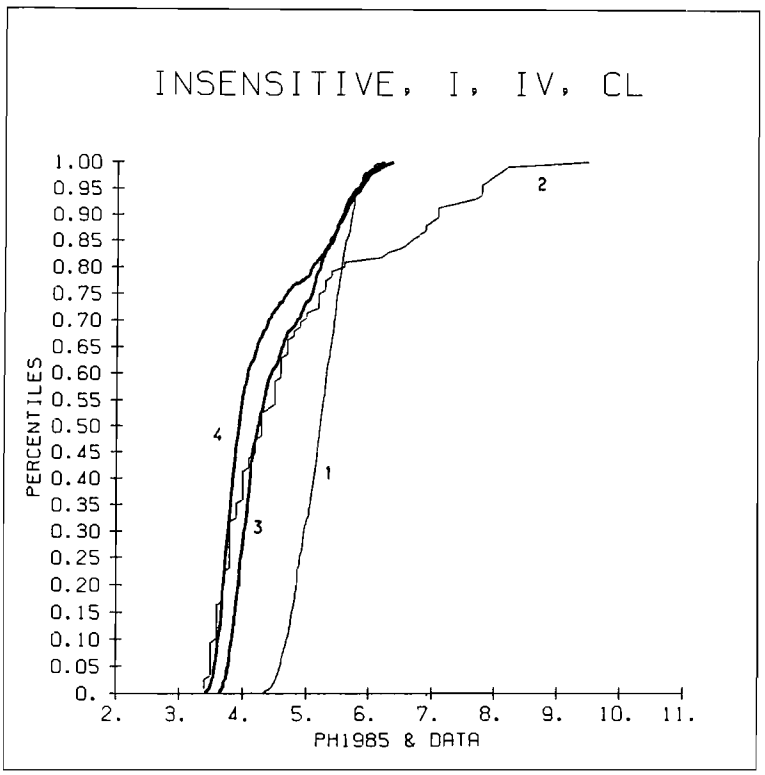

Figure 6.6 Cumulative distributions of the simulated pH in the insensitive class in 1985 due to the Critical Load Acidification (graph 1), to the lowest deposition range (graph 3) , to the highest deposition range (graph 4), compared to the cumulative distributions of the measurements (graph 2).

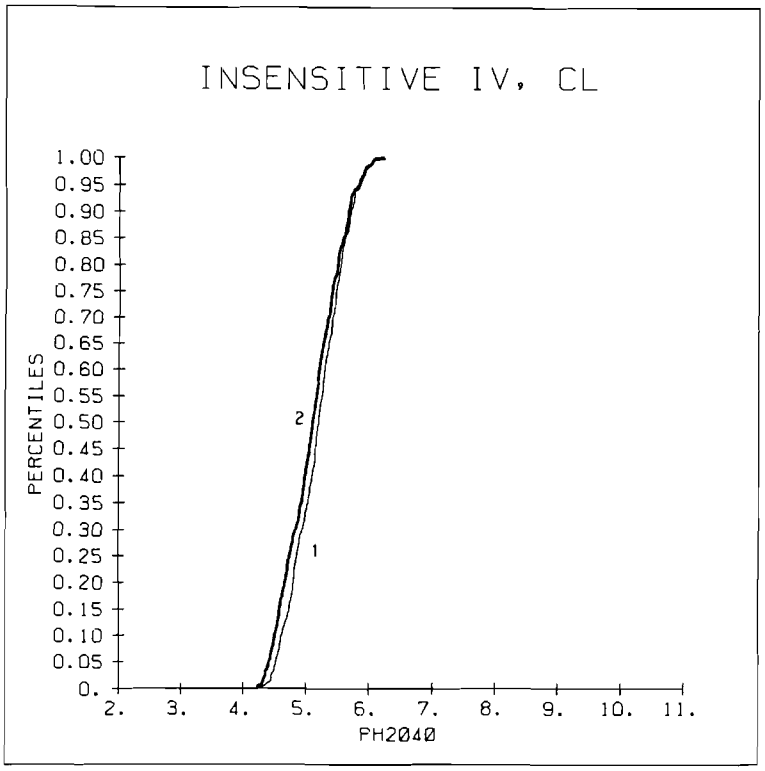

Figure 6.7 Cumulative distributions of the simulated pH in 2040 (graph 2) compared to the cumulative distribution of the Critical Load Acidification in 1985 (graph 1).

within the Critical load range in the Netherlands from 1990 to later years. The reason for this exercise is to estimate the predicted time at which watershed acidification 'recovers' to the Critical Load Acidification of 1985. The result in the insensitive zone is shown in Figure 6.7. The displayed difference between the cumulative distribution of the Critical Load Acidification (graph 1) and the 'recovery' pH (graph 2) occurs after 2015 and hardly changes in later years. Note that the closeness of the distribution functions leads to rather large differences in the percentages of lakes having reached a particular acidification level, e.g. about 30% of the lakes have a pH smaller or equal to 5.0 in the Critical Load Acidification case (Figure 6.7, graph 1) against about 40% in the 'recovery' case (Figure 6.7, graph 2). An exact overlay of the Critical Load Acidification distribution with the 'recovery' distribution is obtained in 2005 for catchments in the sensitive zone.

Finally the influence of buffer processes in the soils were tested by enforcing the model to use the whole Cation Exchange Capacity in stead of a fraction (base saturation; see chapter 3). The results of this analysis did not exhibit much difference with the results described above. This suggest that models in which soil processes are less relevant may be appropriate as well for the policy planning of surface water quality in the Netherlands by means of flexible zoning.

6.4 CONCLUDING REMARKS

In the foregoing chapter the RAINS Lake Model was applied to Dutch lakes and moorland-pools. This exercise was useful because data were available for the soil and lake characteristics for the Netherlands as a whole, rather than for a number of predetermined regions. It could be shown that the method of flexible zoning leads to a partitioning of Dutch catchments into zones with distinct characteristics.

The Netherlands were partitioned into four deposition classes that cover the North to South deposition pattern of the country, i.e. (1) 3600-4400, (2) 4400-5200, (3) 5200-6000, (4) 6000-6800 mol/ha/yr of potential acid. Catchment characteristics that are sensitive and insensitive to acid deposition were defined by partitioning the ranges

of the most important parameters. These important parameters explain most of the variance of predicted pH in 1985 (see chapter 2). The combination of four deposition classes and two sensitivity classes gave a total of eight zones for the Netherlands. A series of model simulations were performed from 1900 to 1985 with deposition levels fixed at the critical loads or to one of the four deposition classes. Predictions after 85 years of lake acidification where then compared to measurements. It was shown that the RAINS Lake Model, when applied to Dutch inputs, simulates a cumulative pH in the insensitive zone of the first deposition class that is similar to about the 70th percentile of the cumulative distribution of the measurements. For the Dutch region as a whole, that is the aggregate result without zoning, only predictions below the lower 30th percentile are reasonably represented. However the effect of acidification from historical and future critical load deposition are predicted differently in the sensitive and insensitive zones. The acidification due to a 1900 to 1985 deposition of critical loads leads to a Critical Load Acidification in 1985 of a pH near 4 in the sensitive zones and between a pH of 4 and 6 in the insensitive zones. It is shown that when the critical loads are deposited starting in 1990, the RAINS Lake Model will predict the Critical Load Acidification will be achieved after a minimum of 15 years.

In reality it will be difficult to achieve a Critical Load Acidification level by 1990 because other countries must collaborate to reduce total emissions. Furthermore, the model predictions were obtained using only one model, i.e. the RAINS Lake Model. This study did not investigate and can not exclude whether another model would show different results. It has been shown, however, that the method of flexible zoning can be implemented even when data are available on an interval scale. It is, therefore, recommended that different models be used with the flexible zoning procedure. Data monitoring programs should also be designed to obtain the information that allows for models to be run with Monte Carlo based methods like flexible zoning. Once flexible zoning has been applied, a policy maker can use the predictions of the sensitive watersheds to estimate the frequency of lakes within any (bounded) region that will be at risk. It is recommended that the quality of the data be improved as simulation results become available. This was illustrated in this chapter by the

differences in watershed parameter intervals that lead to different model predictions in sensitive and insensitive zones. The simultaneous improvement in models and data may increase the speed with which policy makers may recommend remedial action.

Model predictions and resulting policy measures may become more targeted when Dutch catchments are flexibly zoned as a function of catchment and deposition properties.

7. CONCLUSION

7.1 REFLECTION ON THE STUDY

The scale at which human activities have interacted with our natural environment has grown over the past decades. The complexity of these interactions has also increased. Therefore, it is now difficult to recognize and isolate particular human activities so that the quality of our environment can be improved. Climatic change, for example, is due to a great many independent human activities operating at different temporal and spatial scales.

Policies aimed at the alleviation of negative environmental impacts have increasingly been based on predictions made with artificial representations, i.e. models of the systems involved. The usefulness of these models, however, are limited by many uncertainties, e.g. (1) is the complexity of the system properly reflected in the model structure? (2) are the data used in the model representative of the system? (3) is the temporal and spatial scale used in the model appropriate to understand the system's behavior?

These and other issues related to systems and modeling in general and environmental modeling in particular constitute the context of chapter 1 and 2 of this study. Chapters 3 through 6 are more technical in nature and concentrate on the modeling of watershed acidification. The purpose of the study is to:

(1) provide a method of partitioning the spatial scale of model predictions in order to increase the confidence in the predictive performance.
(2) describe the relationship between regional characteristics and the level of detail at which system processes are represented in the model.
(3) implement the concept of critical loads to provide policy insight concerning surface water quality on broad spatial scales.

Many methodological issues were encountered in the course of the investigations. These include:

- the relationship between system's perception and model complexity,
- models with different solution methods, levels of detail, integration of different processes, inclusion of time, and inclusion of stochastic techniques,
- different stages in model building,
- different structures applied in Integrated Environmental Modeling,
- the treatment of uncertainty,
- the relation between uncertainty and sampling techniques,
- methods for calibrating model predictions to represent particular characteristics of a system,
- processes involved in watershed acidification, and
- the relationship between model predictions of watershed acidification and the use of a spatial zoning system.

Chapter 1 provides an introductory treatment to issues of systems analysis and modeling, e.g. system's perception, model complexity, model uncertainty and policy applicability. In the analysis of systems the use of simulation models and probabilistic methods, have become common for understanding systems, determining the aggregation level of subsystems, defining their interactions, uncertainties and data requirements. This study elaborates on the relation between uncertainty, model complexity and data aggregation in the field of environmental modeling in general, and the modeling of watershed acidification in particular.

Chapter 2 provides an overview of Integrated Environmental Model (EIM) structures and describes some examples of EIM's. One of these models is the Regional Acidification INformation and Simulation (RAINS) model that predicts the pollution of sulfur and nitrogen oxides on the European scale from 1960 to 2040. RAINS provides its users with the possibility of evaluating sulfur and nitrogen abatement strategies that can interactively be related to fuel combustion. Evaluation of RAINS strategies may consist of investigating sulfur and nitrogen deposition patterns over Europe, of forest soil acidification, of direct forest

impacts from sulfur dioxide concentrations and of lake acidification. The latter module is called the RAINS Lake Model which has been used in the technical parts of this study. The RAINS Lake Model has previously been applied to 14 predefined regions in Scandinavia, i.e. 5 regions in Finland, 6 regions in Sweden and 3 regions in Norway. Data about the soil and catchment characteristics, necessary for model simulations, are available in the form of value ranges and frequency distributions for each of these regions. The acidity, i.e. the pH of the lakes in each region has been sampled as well. In chapter 2 attention is given to the way in which the RAINS Lake Model has been calibrated to represent these pH measurements. The calibration result is used in the RAINS model to predict the state of lake acidification that results from a change of acid deposition due to abatement policies in Europe. The potential of the RAINS model for the evaluation of policy alternatives has been recognized within the Economic Commission for Europe (ECE). Chapter 2 finally addresses the technical aspects of uncertainty in model structure, parameter estimates, observed inputs and model outputs. Methods to quantify uncertainty are presented including a computer package, PRISM, that integrates the sampling from model parameter ranges with the execution of the model and the statistical analysis of the resulting set of predictions. PRISM is used in this study to perform Monte Carlo Simulation on the RAINS Lake Model with the purpose of (1) defining the most important Monte Carlo parameters of the RAINS Lake Model accounting for the variability of pH predictions, and (2) testing the effect of calibration on the quality of the RAINS Lake Model in the 14 Scandinavian regions mentioned above.

Chapter 3 consists of an overview of chemical processes involved in the acidification of soils and surface waters due to acid deposition. The simulation of the different levels of detail by which these processes are represented in four different lake acidification models including RAINS is discussed. The chapter also describes the equations of the RAINS Lake Model.

Chapter 4 elaborates on the definition of a region, its boundaries and the relationship with regional model predictions. The objective of the chapter is to determine the effect that predefined regional boundaries have on the estimate of broad scale watershed acidification.

Two kinds of models are used (1) the RAINS Lake model and (2) a simple model, i.e. a metamodel, relating RAINS Lake model predictions to its most important parameters. PRISM is used to calibrate both models. The RAINS Lake Model is calibrated in each of the southern regions in Scandinavia, by adjusting the value ranges of the most important parameters. Next, the difference of the model predictions to regional measurements is evaluated when (1) the calibration result is used in the other regions within the same country (intra country extrapolation) and (2) when the calibration result including the value ranges of other regional characteristics in the southern region of Finland are used in all the other regions (inter country extrapolation). The metamodel is calibrated in the southern region of Finland and its predictions then tested by using deposition ranges and regional characteristics different from the ones used for the calibration. Results of both model analyses show that changing combinations of deposition levels and regional characteristics may (1) lead to shifts in the ranking of model parameters with respect to their importance in explaining the uncertainty of model predictions, and (2) affect the quality of model predictions. The conclusions following from the analysis are:

- calibration of models to predefined regions may be of limited use for policy purposes because predictions of environmental effects (i.e. watershed acidification) as result of changing deposition patterns over large regions may be error prone.
- Simple models may replace more complex models if the types of watersheds are similar with respect to their response to acid deposition.

Finally chapter 4 notes that similarity exists between these results and the findings reported in the field of spatial interaction modeling.

Chapter 5 introduces an alternative method for obtaining model predictions as a function of model calibration within predefined regions. This method is called the 'method of flexible zoning' and consists of:

(1) applying uncertainty analysis using Monte Carlo Simulation on the Monte Carlo parameters of the model as applied to an arbitrary bounded region,

(2) subdividing the ranges of the most important parameters, found

in (1), to allow for different types of soil and watershed characteristics. Combinations of subranges of different parameters can be grouped into for zones of watersheds within a region that react in a similar way to acid deposition (i.e. 'sensitive' or 'insensitive' watersheds),

(3) performing Monte Carlo Simulation on each of these zones subject to different levels of acid deposition,

(4) investigating the homogeneity of the watershed characteristics in each zone by regression analysis (i.e. developing a metamodel),

(5) applying the cumulative distributions of model predictions to assess policy intentions, e.g. target loads of acid deposition.

The percentage of lakes is determined of which the acidification, which results from this target load, may still lead to harmful effects to flora and fauna. The highest deposition at which no long term harmful effects occur, i.e. the critical load, may be found to vary between zones.

The method is illustrated for the southern region in Norway of which the most important parameters are partitioned with values extracted from the literature. The results show that zones, constructed by the method of flexible zoning, may need to be partitioned again to obtain similar kinds of watersheds in each zone. Finally it is shown that the assessment of a critical pH leads to different percentages of lakes at risk in each zone.

Chapter 6 consists of an application of the method of flexible zoning to the Netherlands. Four zones of acid deposition were distinguished, i.e. 3600-4400, 4400-5200, 5200-6000 and 6000-6800 mol/ha thus covering the entire Dutch area. The combination of these deposition ranges with 'sensitive' and 'insensitive' partitionings of the most important Monte Carlo Simulation parameters in the Netherlands (a total of 8 zones) led to satisfactory metamodel simulations. Thus there was no need to subdivide these zones. The RAINS Lake Model was applied in each zone over the period from 1900 to 1985 and from 1900 to 2040. Field measurements were available for 1984. By applying a critical load range of 250-800 mol/ha/yr starting from 1900, a state of acidification in 1985 was simulated for each zone. This state of acidification is referred to as the Critical Load Acidification.

Critical Load Acidification was compared to RAINS Lake Model predictions in every zone. It was shown that different predictions were obtained when using the RAINS Lake Model in 'sensitive' and 'insensitive' zones. The cumulative distribution of the pH predictions in the insensitive zone due to a range of depositions between 3600 and 4400 mol/ha showed resemblance, below the 70 percentile, with the cumulative distribution of the measurements in 1984. Finally the RAINS Lake Model predicted that it would take at least 15 years to reach the Critical Load Acidification in the Netherlands if depositions would be reduced to the critical load range after 1990.

7.2 OUTLOOK

In order to improve the understanding of dose response relationships, monitoring programs should cover a sufficiently large temporal and spatial scale to be able to quantify the decrease as well as the recovery of the state of environmental systems. Furthermore, models should continue to be developed to describe the dose response relationships. These models should be capable of predicting the state of a system that was documented by the monitoring program.

Complete compatibility between models and data is uncommon. First, the temporal and spatial scales at which changes occur in the quality of the environment are different for many processes in the natural environment. As pointed out in chapter 1, it is possible to view systems in a holistic or reductionistic way. Models developed within the frame of each of these views will be different with different data needs. The different kinds of Integrated Environmental Models described in chapter 2 illustrate this point. Second, the measurements do not necessarily provide information about the system elements that lead to the observed phenomena. Third, the measurements are not necessarily suited for the model to reproduce the observed phenomena. The global phenomena that result from many interactions between human activities and environmental resources may not allow for enough time needed to obtain 'better' data and 'better' models. In fact it is questionable whether consensus will ever be reached about the quality of models and data needed to understand and reverse the depletion of the

environmental quality. For example, the depletion of the quality of surface waters has been simulated by several models (e.g., ILWAS, ETD, MAGIC and RAINS) as described in chapter 3. Each of these models differ from one another in the level of detail at which similar processes are simulated. This distinction between models makes it necessary to use calibration procedures, especially when model predictions apply to a region rather than a single watershed. Chapter 4 has illustrated how calibration procedures may affect model predictions. It is, therefore, questionable whether predictions of different models which simulate similar processes can be brought under a common denominator by choosing the 'right' calibration procedure.

With the increasing importance of integrated environmental modeling in combination with the analysis of uncertainty of model predictions a complete consensus may not be needed. Uncertainty analysis of a model which integrates different processes (i.e., meteorological, soil and watershed acidification processes) has been shown in chapter 5 and 6 to allow for ranges of model predictions, i.e., cumulative distributions for different combinations of watershed characteristics. Such a probabilistic interpretation of model predictions in combination with risk assessments of environmental effects may allow different models and data to provide overlapping confidence intervals around predictions of environmental state variables.

The method of flexible zoning introduced in this study allows for a probabilistic investigation of the compatibility between models and available spatial data. The application of a wide set of statistical techniques, including the method of flexible zoning, should be further investigated as a means to increase the understanding and the confidence in policy measures taken to reverse the depletion of the quality of our natural environment.

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SAMENVATTING

De aantasting van ons natuurlijk milieu manifesteert zich op tal van manieren variërend van lokale problemen als geluid en stankoverlast tot grootschalige veranderingen in het klimaat. Het brede scala van milieuproblemen heeft geleid tot de behoefte om het systeem van oorzakelijke verbanden tussen menselijke activiteiten en de vermindering van de kwaliteit van het milieu nader te onderzoeken.
Dit heeft de ontwikkeling van Geïntegreerde Milieu Modellen tot gevolg gehad waarin door middel van wiskundige formuleringen meerdere systemen en hun onderlinge samenhang worden beschreven. De voorspellingen die met deze modellen kunnen worden gemaakt over de kwaliteit van onderdelen van het milieu, zijn voor beleidsmakers interessant omdat op deze wijze diverse beleidsopties met elkaar kunnen worden vergeleken. Deze voorspellingen zijn evenwel onderhevig aan tal van onzekerheden die het gevolg kunnen zijn van onder andere (1) een onjuiste perceptie van de complexiteit van het systeem dat moet worden gemodelleerd, (2) het niet representatieve karakter van de data en (3) een onvoldoende weergave van de temporele en ruimtelijke schaal waarbinnen systeemveranderingen zich voordoen.

Deze en andere onderwerpen met betrekking tot systemen en modellen in het algemeen en milieumodellen in het bijzonder komen aan de orde in de hoofdstukken 1 en 2 van deze studie. De theoretische nadruk van deze twee hoofdstukken contrasteert met de meer technisch georienteerde hoofdstukken 3 tot en met 6. De inhoud van deze hoofdstukken concentreert zich op de modellering van de kwaliteit van het oppervlaktewater als gevolg van zwavel en stikstof depositie, beter bekend als 'zure regen'.

Het doel van deze studie is:

- de ontwikkeling van een methode om de ruimtelijke schaal van een model onder te verdelen op een wijze die de betrouwbaarheid van modelvoorspellingen verhoogd.
- de relatie te beschrijven tussen regionale karakteristieken en de mate waarin systeemonderdelen in het model zijn gedetailleerd.
- de verhoging van de bruikbaarheid van het concept van kritische

waarden, i.e. de maximale waarde waarbij er geen schade wordt geconstateerd aan onderdelen van het milieu, om beleidsinzicht te verschaffen in de oppervlakte-waterkwaliteit in grote regio's.

Hoofdstuk 1 bevat een inleiding in onderwerpen die van belang zijn voor het begrip van systeemanalyse en modelbouw. Voorbeelden hiervan zijn (1) de wijze waarop systemen kunnen worden geïnterpreteerd (holistisch versus reductionistisch) (2) de gevolgen hiervan voor modelstructuren (3) de keuze van het aggregatieniveau van modellen en data en (4) het doel en gebruik van onzekerheids- en gevoeligheidsanalyse. Termen als "onzekerheid", "modelcomplexiteit", en "aggregatie" vormen sleutelbegrippen in deze studie.

In hoofdstuk 2 wordt de opzet behandeld van geïntegreerde milieumodellen en geeft enkele voorbeelden van deze modellen.
Een van deze modellen is het "Regional Acidification INformation and Simulation" (RAINS) model dat is ontwikkeld aan het "International Institute for Applied Systems Analysis" (IIASA) te Laxenburg in Oostenrijk. RAINS voorspelt de emissies en neerslag van zwavel- en stikstofoxiden in Europa van 1960 tot 2040 als gevolg van de verbranding van energiedragers. In dit model kunnen emissie-verminderingen worden gedefinieerd, en de effecten van dergelijke verminderingen of combinaties van verminderingen kunnen worden geëvalueerd in termen van (1) depositie (2) bodemverzuring (3) direkte schade aan bossen en (4) verzuring van oppervlakte wateren in noord Europa. Het laatst genoemde onderdeel, het RAINS Lake Model, is toegepast op 14 regios in Scandinavië, te weten 5 in Finland, 6 in Zweden en 3 in Noorwegen. Gegevens over karakteristieken van de bodem en het watergebied evenals de verzuringsgraad (pH) zijn voor elke regio beschikbaar in de vorm van intervalwaarden en statistische dichtheidsfuncties. Hoofdstuk 2 schenkt verder aandacht aan de wijze waarop het RAINS Lake Model wordt gecalibreerd aan de gemeten pH waarden. De technische aspecten van calibratie en het hiermede samenhangende gebruik van random generatoren en Monte Carlo Simulatie krijgt tevens uitgebreide aandacht. Dit hoofdstuk gaat ook in op methoden om onzekerheidsanalyses toe te passen, zoals het computer-programma, PRISM, dat is ontwikkeld aan het "Oak Ridge National Laboratory" (ORNL) in de Verenigde Staten. PRISM integreert (a) de

herhaalde a-selecte trekking (volgens de "Latin Hypercube Methode") uit intervallen van waarden die worden toegekend aan de inputs van het model (b) de executie van het model, en (c) de statistische analyse van de modelvoorspellingen. PRISM werd in dit onderzoek gebruikt in combinatie met het RAINS Lake Model om (1) de Monte Carlo parameters te selecteren die het meest verklaren van de variabiliteit van de pH voorspellingen en (2) het effect te evalueren van de calibratie van het RAINS Lake Model in de 14 bovengenoemde regio's.

Hoofdstuk 3 geeft een overzicht van de chemische processen die bijdragen tot de verzuring van de bodem en van de oppervlaktewateren. Tevens zijn de wiskundige vergelijkingen beschreven die in het RAINS Lake Model zijn opgenomen om de genoemde processen te simuleren.

In hoofdstuk 4 wordt de relatie uitgewerkt tussen model-voorspellingen per regio en de wijze waarop een gebied wordt opgedeeld. Het doel van dit hoofdstuk is de betrouwbaarheid van de modelvoorspellingen van de waterkwaliteit te evalueren als functie van strikt begrensde ruimtelijke gebieden. Twee modellen met een verschillende graad van complexiteit worden toegepast: (1) het RAINS Lake Model en (2) een vereenvoudiging van het RAINS Lake Model, i.e. een metamodel, waarbij de pH-voorspellingen worden verklaard door de meest belangrijke Monte Carlo parameters van het RAINS Lake Model. Beide modellen worden gecalibreerd met behulp van PRISM. De calibratie bestaat uit het aanpassen van de verdelingen van de meest belangrijke parameters. Dit gebeurt met het eerste model voor de zuidelijke regio in elk van de drie landen, terwijl het metamodel alleen in de zuidelijke regio van Finland wordt gecalibreerd. Het verband tussen regionale condities, met inbegrip van zwavel depositiepatronen, en voorspellingen van het RAINS Lake Model wordt als volgt geëvalueerd: (a) het calibratieresultaat, in elk van de zuidelijke regio, wordt toegepast in de andere regio van hetzelfde land (intra-extrapolatie) en (b) het calibratieresultaat met inbegrip van de waarden van de overige parameters in de zuidelijke regio van Finland wordt toegepast in alle overige regio's (inter-extrapolatie). De voorspellingen van het metamodel als functie van regionale condities worden geëvalueerd door het verkregen calibratieresultaat toe te passen in de Noordelijke regio van Finland. Tevens worden voorspellingen van het metamodel voortvloeiend uit hoge deposities vergeleken met de voorspellingen van het RAINS Lake Model onder dezelfde condities.

De resultaten van beide modelanalyses geven aan dat veranderende combinaties van regionale omstandigheden en depositiepatronen leiden tot: (1) een verschuiving van de volgorde in de belangrijkheid van de Monte Carlo parameters met betrekking tot de verklaring van de variantie van de pH-voorspellingen, en (2) een aantasting van de kwaliteit van de regionale modelvoorspellingen. Tevens wordt geconstateerd dat deze resultaten overeenkomst vertonen met bevindingen in de discipline van de ruimtelijke graviteitsmodellen. De conclusies die uit de analyses van hoofdstuk 4 worden getrokken, zijn:

- de modelvoorspellingen die nodig zijn voor de evaluatie van de waterkwaliteit in grote regio's als gevolg van veranderende condities (regio's, depositie) kunnen aan betrouwbaarheid inboeten als gevolg van de inconsistentie tussen het calibratieresultaat en deze condities.
- Eenvoudige (meta-) modellen kunnen meer complexe modellen vervangen als de oppervlakte wateren vergelijkbaar zijn, m.a.w. homogene karakteristieken vertonen, met betrekking tot hun gevoeligheid voor zure depositie.

In hoofdstuk 5 wordt een alternatief gepresenteerd om de in het voorgaande hoofdstuk geconstateerde tekortkomingen van regionale voorspellingen van de milieukwaliteit te compenseren. Deze "flexibele zoneringsmethode" bestaat uit de volgende stappen:

(1) de toepassing van onzekerheidsanalyse op het model in een strikt begrensd gebied,
(2) de onderverdeling van de waarde-intervallen van de meest belangrijke parameters, volgend uit (1), teneinde verschillende groepen van karakteristieken met betrekking tot de bodem en het watergebied te kunnen onderscheiden. Combinaties van deelintervallen van verschillende parameters vertegenwoordigen verschillende zones van watergebieden die "gevoelig" of "ongevoelig" zijn voor zure depositie.
(3) de toepassing van Monte Carlo Simulatie op het model in elk van deze zones onder een variërende exogene invloed (zure depositie).
(4) de toepassing van metamodellen om inzicht te krijgen in de mate van homogeniteit van de oppervlaktewateren in de zones.
(5) de evaluatie van emissieverminderingen of andere (beleids-)

maatregelen met betrekking tot nagestreefde depositiewaarden ("target levels") aan de hand van de cumulatieve verdeling van de pH voorspellingen in elke zone.

Het blijkt dat de kritische waarde kan variëren voor elke zone. De methode van flexibele zonering werd, in hoofdstuk 5, geïllustreerd in de zuidelijke regio van Noorwegen waarbij de spreiding van de meest belangrijke parameters tevens zijn bewerkt aan de hand van de bestaande literatuur.

Hoofdstuk 6 beschrijft de data, de modelaanpassingen en de resultaten van de toepassing van de methode van flexibele zonering met het RAINS Lake Model in Nederland. Er worden vier zure depositie-intervallen in Nederland onderscheiden, resp. 3600-4400, 4400-5200, 5200-6000 en 6000-6800 mol/ha/jaar. Met deze depositie-intervallen is het gehele Nederlandse grondgebied afgedekt. Deze depositie intervallen worden gecombineerd met gevoelige en ongevoelige waarden van de meest belangrijke parameters (bijvoorbeeld, respectievelijk, dunne en dikke bodems). Deze combinaties heten "zones". Het RAINS Lake Model wordt in elke zone toegepast in de perioden 1900-1985 en 1900-2040. Veldmetingen uit 1984 worden in het onderzoek vergeleken met de modelvoorspellingen. Door de toepassing van kritische depositiewaarden uit het interval van 250-800 mol/ha/jaar, m.a.w. de maximale waarden waarbij in brede kring wordt aangenomen dat er geen schade aan flora en fauna wordt toegebracht, wordt een staat van verzuring in 1985 voorspeld. Deze "Kritische Waarde Verzuring" is gebruikt als referentie waartegen andere modelvoorspellingen kunnen worden afgezet.

Er is aangetoond dat modelvoorspellingen verschillen wanneer het RAINS Lake Model wordt toegepast op gevoelige en ongevoelige zones. Verder blijkt de cumulatieve verdeling van de voorspellingen in de ongevoelige zone bij het depositie interval 3600-4400 mol/ha, tot ca. het 70 percentiel overeen te komen met de cumulatieve verdeling van de metingen. Gegevens ontbreken om, in het kader van deze studie, de karakteristieken van de watergebied-intervallen te valideren die tot deze modelvoorspelling leiden.

Tenslotte voorspelt het RAINS Lake Model dat het minstens 15 jaar duurt alvorens de Kritische Waarde Verzuring wordt bereikt wanneer met ingang van 1990 de depositie in Nederland zou worden teruggebracht tot

de kritische depositiewaarden.

In het algemeen kan worden geconcludeerd dat de methode van flexibele zonering de betrouwbaarheid van modelvoorspellingen verhoogt in vergelijking tot voorspellingen die zijn gebaseerd op modelcalibratie. De reden is dat de methode het mogelijk maakt om de compatibiliteit tussen het model en regionale condities te variëren. Gegevens kunnen worden verzameld om informatie te krijgen die de toepassing van flexibele zonering op modellen mogelijk maakt. Op basis van de modelvoorspellingen over gevoelige zones die met de methode van flexibele zonering worden verkregen kan de beleidsmaker de frequentie schatten van meren in een afgebakende regio die een verhoogd verzuringsrisico dragen. De methode van flexibele zonering verschaft derhalve inzicht in de combinaties van regionale condities en exogene invloeden (depositie) die leiden tot veranderingen in de onzekerheid van modelvoorspellingen.

CURRICULUM VITAE

Jean-Paul Hettelingh werd op 22 februari 1954 in Amsterdam geboren. In 1972 behaalde hij het diploma HBS-B aan het Snellius Lyceum (tegenwoordig Snellius Scholen gemeenschap) te Amstelveen en studeerde van 1972 tot 1981 econometrie aan de Interfaculteit voor Actuariële Wetenschappen en Econometrie van de Vrije Universiteit te Amsterdam. Het kandidaatsexamen econometrie werd op 26 februari 1976 afgelegd en het doctoraalexamen werd op 29 januari 1981 behaald.

In de periode van 1978 tot 1981 vervulde hij diverse student-assistentschappen aan het Instituut voor Milievraagstukken (IvM) van de Vrije Universiteit in welke functie hij deelnam in de statistische, modelmatige en computerkundige aspecten van diverse projecten. In de periode van 1981 tot 1985 zette hij tal van projectgerichte activiteiten binnen het IvM voort in de hoedanigheid van wetenschappelijk ambtenaar.

In oktober 1985 volgde de aanstelling tot research scholar aan het International Institute for Applied Systems Analysis (IIASA) te Laxenburg in Oostenrijk. Tot juni 1989 werkte hij daar aan het RAINS model voor de beschrijving en simulatie van verzuring in Europa. Een aantal hieruit voortvloeiende activiteiten werden deels op IIASA en deels aan het Oak Ridge National Laboratory in de Verenigde Staten volbracht. Veel van de resultaten van deze onderzoeksonderdelen maken deel uit van dit proefschrift.

Thans is hij hoofd van de Afdeling Modelonderzoek en Modelbeheer van het Laboratorium voor Afvalstoffen en Emissies aan het Rijks Instituut voor Volksgezondheid en Milieuhygiëne (RIVM) te Bilthoven.

Hij is de echtgenoot van Elisabeth Jacqueline Garnier en de vader van Charlotte Elodie Alix.

APPENDIX I

Appendix I presents an efficient algorithm for the calibration 'filtering' procedure of the RAINS Lake Module (Posch, 1987[1]; appendix of Sutton, 1987).

The description of the aim of the filtering procedure can be found in chapter 2.

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[1]Used with permission of Dr. Maximilian Posch.

The aim of this note is to derive an optimal procedure (filter) for fitting Monte Carlo simulations to an observed (measured) cumulative distribution function (cdf).

The cdf of the observations is characterized by values $y_0 = 0$, $y_1, \ldots, y_{n-1}$, $y_n = 1$ (see Figure 1) and is derived from a histogram with n bins.

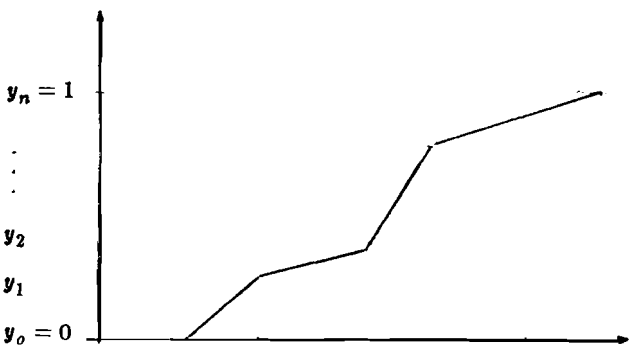

Figure 1: One-dimensional cdf for a histogram.

Let N_j be the number of simulations with outcome falling into the jth bin. The aim is to derive a formula for the maximal number of *accepted runs* per bin, a_j $(j = 1,\ldots,n)$, in such a way, that -- after rejection of $N_j - a_j$ runs per bin -- the simulated cdf and the observed cdf are the same. This requires that

$$
\begin{aligned}
y_1 &= \frac{a_1}{a} \\
y_2 &= \frac{a_1 + a_2}{a} \\
y_3 &= \frac{a_1 + a_2 + a_3}{a} \\
&\cdots\cdots\cdots \\
y_n &= \frac{a_1 + \cdots + a_n}{a}
\end{aligned}
\tag{1}
$$

where $a := \sum_{j=1}^{n} a_j$ is the total number of accepted simulations. These are n equations for the n unknowns $a_1, \ldots, a_n$. However, the last equation in (1) is an identity $(1 = 1)$, so that we have only $n-1$ equations. This means that we have an additional degree of freedom to

fix $a_1,\ldots,a_n$. It is clear from the definitions that we have the following constraints on the a_j's

$$0 \leq a_j \leq N_j \ , \quad j=1,\ldots,n \tag{2}$$

Next we subtract the $(j-1)$th equation from the jth equation in (1) and get after rearranging

$$x_j := \frac{a_j}{y_j - y_{j-1}} = a \ , \quad j=1,\ldots,n \tag{3}$$

where we have introduced the new unknowns x_j. (3) can be also written as

$$x_1 = x_2 = \cdots = x_n = a \, , \tag{4}$$

showing that all the x_j's are identical to the total number of accepted runs. The constraints for the x_j's read [compare (2)]

$$0 \leq x_j \leq \frac{N_j}{y_j - y_{j-1}} \ , \quad j=1,\ldots,n \tag{5}$$

Now let M be the minimum of the upper bounds for the x_j:

$$M := \min\{ \frac{N_j}{y_j - y_{j-1}}, j=1,\ldots,n\} \tag{6}$$

Then it follows from (4) that

$$0 \leq x_j \leq M \quad \text{for all } j \tag{7}$$

Now we fix the additional degree of freedom of the a_j's in such a way that we request the a_j's (the number of accepted runs) to be maximal. From the definition of the x_j's [see (3)] it follows that the x_j's have to be maximal, and from (7) it follows that

$$x_j = M \quad \text{for all } j \tag{8}$$

which means that the optimal a_j's are given by [see (3)]

$$a_j = (y_j - y_{j-1})M \ , \quad j=1,\ldots,n \tag{9}$$

with M given by (6).

The generalization to more dimensions is straightforward and mainly a matter of notation and proper interpretation of the one-dimensional result.

We note that $y_j - y_{j-1} =: \Delta y_j$ is nothing else but the number of observations in bin j divided by the total number of observations ($\sum_{j=1}^{n} \Delta y_j = 1$). Therefore we get the following result for the d-dimensional case:

The maximal number of accepted simulations for bin $(j_1,\ldots,j_d)$ is given by

$$a_{j_1,..,j_d} = \Delta y_{j_1,..,j_d} M \tag{10}$$

where

$$M := \min\{ \frac{N_{j_1,..,j_d}}{\Delta y_{j_1,..,j_d}}, j_k = 1,..,n_k, k = 1,...,d\} \tag{11}$$

Here $N_{j_1,..,j_d}$ denotes the number of simulations for bin $(j_1,..,j_d)$ and n_k is the number of classes of variable k.

A P P E N D I X II

In this appendix a description is given of the PRISM computer code used in this study and of the input file (data.dat) containing the statistics (mean, standard deviation, lower and upper bound) of the parameters that are subjected to Monte Carlo Simulation of the model.

PRISM consists of three blocks:

(1) PRISM1 contains a great number of routines for sampling values from cumulative distributions (e.g. uniform, triangular, normal, etc.). The choice of the type of cumulative distribution, the value-range, the mean and the standard deviation are defined by the inputfile to PRISM1 (data.dat). The latter inputfile contains (1) the number of iterations to be performed with the model, (2) the model parameters that are used in the Monte Carlo Simulation (3) the type of distribution from which values should be assigned to the model parameter and (4) the mean, the standard deviation and the lower and upper bound of the parameter values. The output of PRISM1 is a file (prism1.dat) that contains N (N = the number of Monte Carlo runs) blocks of values that have been sampled for every model parameter. The parameters implemented in this study consisted of soil, watershed and deposition characteristics.

(2) PRISM2, as applied in this study, contains the RAINS LAKE Model. PRISM2 reads prism1.dat (see above) and performs a model run for each of the N blocks of parameter values. The output of PRISM2 is a file (prism2.dat) that contains N blocks of model predictions, i.e. the predicted pH for every year.

(3) PRISM3 is a set of routines computing different statistics (e.g. partial R^2, F-statistic, total, regression and error sum of squares etc.), and percentiles of the pH predictions stored in prism2.dat.

The implementation in PRISM2 of the RAINS Lake Model in general and for the Dutch case in particular and many of the routines of PRISM3 used for this study have been written by the author. The code as well as the input data files used for this study are available upon request. PRISM has been developed by Dr.R.H.Gardner of the Environmental Sciences Division at the Oak Ridge National Laboratory. Figure 4.2 (chapter 4) summarizes the setup of PRISM.

A P P E N D I X III

Appendix III presents the cumulative distributions of the RAINS Lake Module pH response in 1980 for all the lake classes that have been distinguished with the method of flexible zoning in chapter 5. Distributions and corresponding statistics have been plotted for 4 'sensitive' and 4 'insensitive' zones.

An explanation of the results can be found in chapter 5.

Mean	7.019
Absolute Deviation	0.5288
Standard Deviation	0.7125
Variance	0.5077
Skewness	-1.523
Kurtosis	2.514
Minimum	4.464
2.5% percentile	4.703
25% percentile	6.712
Median	7.198
75% percentile	7.502
97.5% percentile	7.896
Maximum	8.070
Number of Iterations	500

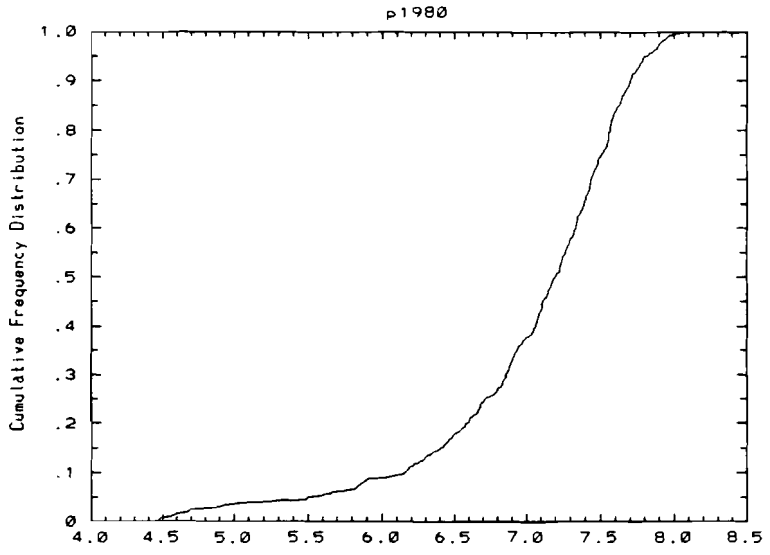

Figure III.1 Cumulative distribution of RAINS Lake Module pH response in 1980 in a sensitive sub-region with deposition of 0 g/m2/yr.

Mean	:	5.829
Absolute Deviation	:	1.368
Standard Deviation	:	1.445
Variance	:	2.089
Skewness	:	-0.3825E-01
Kurtosis	:	-1.741
Minimum	:	4.000
2.5% percentile	:	4.003
25% percentile	:	4.285
Median	:	6.105
75% percentile	:	7.254
97.5% percentile	:	7.791
Maximum	:	7.906
Number of Iterations	:	500

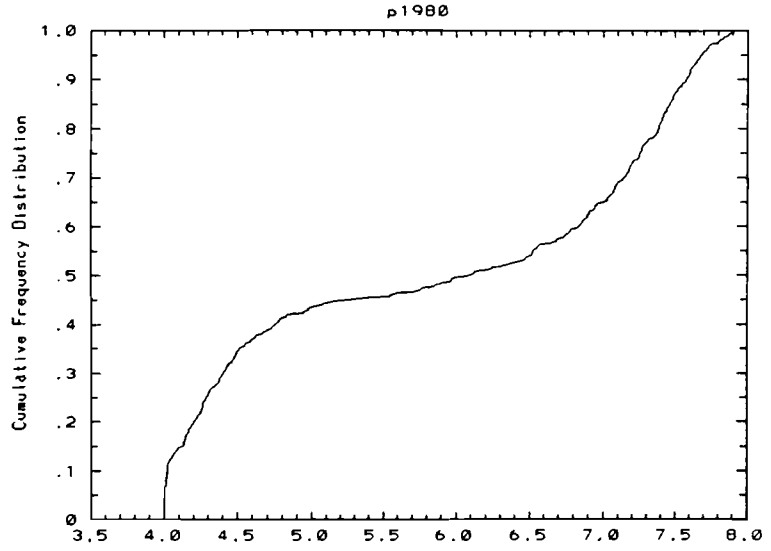

Figure III.2 Cumulative distribution of RAINS Lake Module pH response in 1980 in a sensitive sub-region with deposition of 1 g/m2/yr.

Mean	:	4.719
Absolute Deviation	:	0.9494
Standard Deviation	:	1.180
Variance	:	1.394
Skewness	:	1.380
Kurtosis	:	0.2341
Minimum	:	3.823
2.5% percentile	:	3.901
25% percentile	:	3.977
Median	:	4.058
75% percentile	:	4.699
97.5% percentile	:	7.431
Maximum	:	7.821
Number of Iterations	:	500

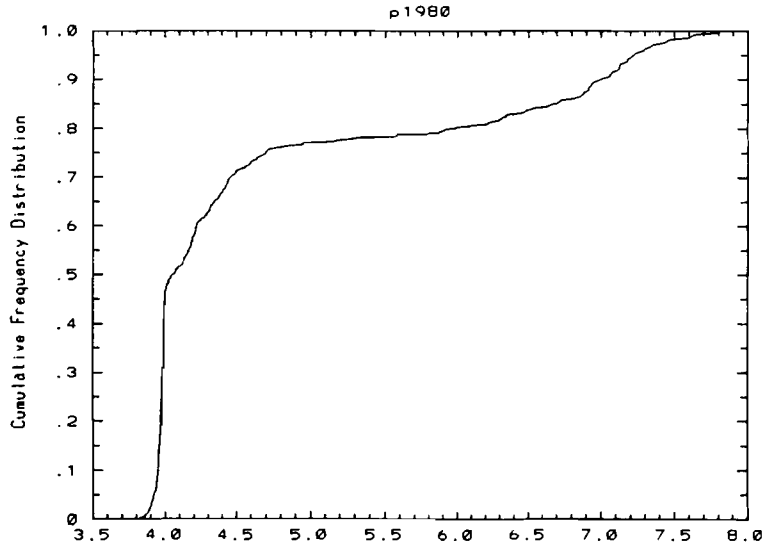

Figure III.3 Cumulative distribution of RAINS Lake Module pH response in 1980 in a sensitive sub-region with deposition of 5 g/m2/yr.

Mean	:	4.477
Absolute Deviation	:	0.8133
Standard Deviation	:	1.081
Variance	:	1.169
Skewness	:	1.696
Kurtosis	:	1.281
Minimum	:	3.583
2.5% percentile	:	3.695
25% percentile	:	3.881
Median	:	3.967
75% percentile	:	4.287
97.5% percentile	:	7.221
Maximum	:	7.527
Number of Iterations	:	500

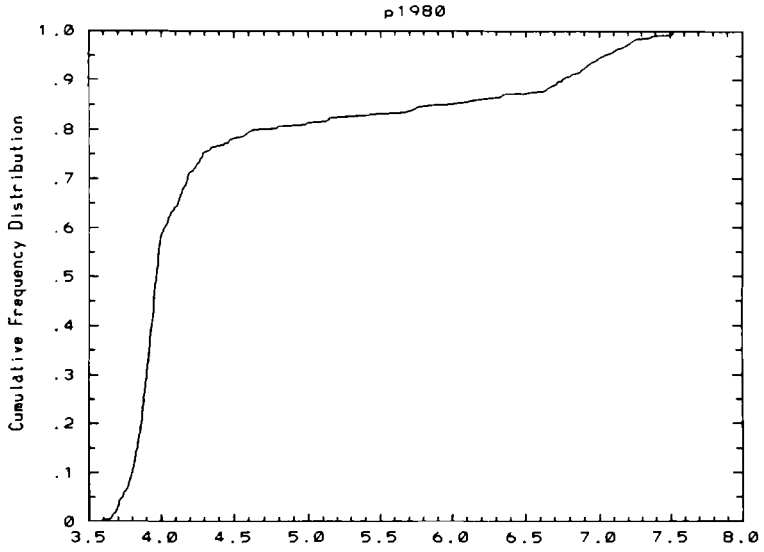

Figure III.4 Cumulative distribution of RAINS Lake Module pH response in 1980 in a sensitive sub-region with deposition of 10 g/m2/yr.

Mean	:	7.627
Absolute Deviation	:	0.1264
Standard Deviation	:	0.1578
Variance	:	0.2489E-01
Skewness	:	0.1403
Kurtosis	:	-0.1615
Minimum	:	7.166
2.5% percentile	:	7.323
25% percentile	:	7.522
Median	:	7.615
75% percentile	:	7.734
97.5% percentile	:	7.948
Maximum	:	8.077
Number of Iterations	:	500

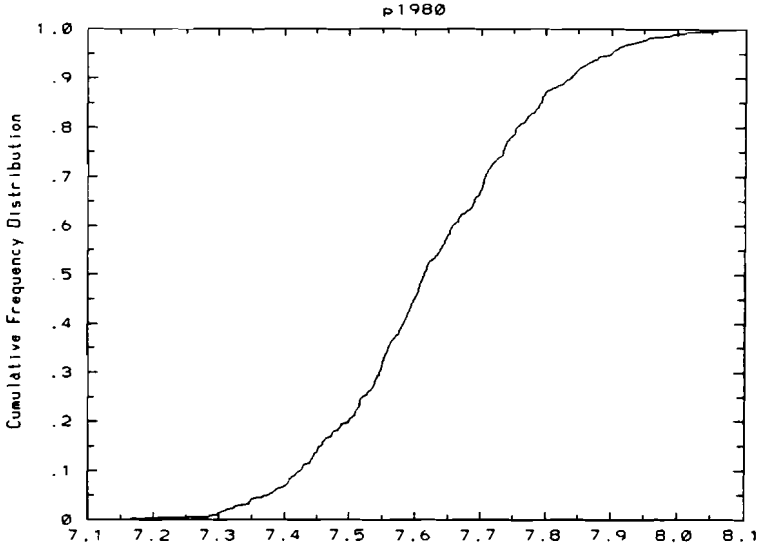

Figure III.5 Cumulative distribution of RAINS Lake Module pH response in 1980 in an insensitive sub-region with deposition of 0 g/m2/yr.

Mean	:	7.468
Absolute Deviation	:	0.1542
Standard Deviation	:	0.1947
Variance	:	0.3790E-01
Skewness	:	-0.4654E-01
Kurtosis	:	-0.1215
Minimum	:	6.902
2.5% percentile	:	7.079
25% percentile	:	7.343
Median	:	7.462
75% percentile	:	7.597
97.5% percentile	:	7.872
Maximum	:	7.930
Number of Iterations	:	500

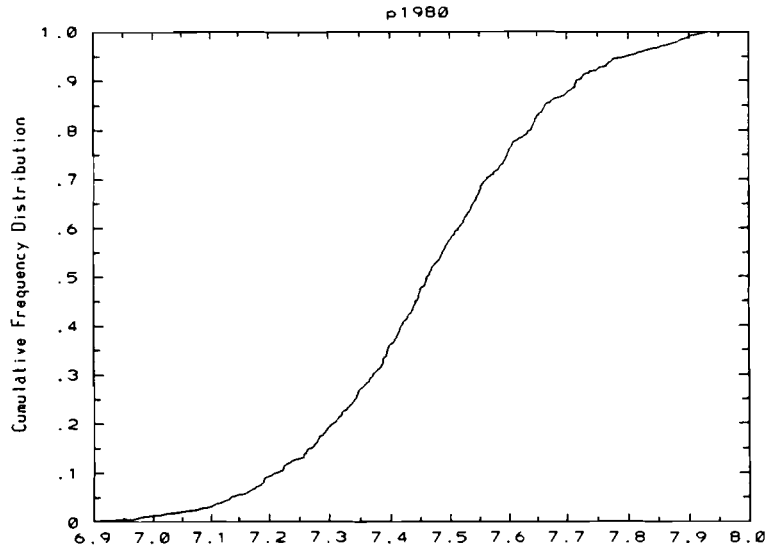

Figure III.6 Cumulative distribution of RAINS Lake Module pH response in 1980 in an insensitive sub-region with deposition of 1 g/m2/yr.

Mean	:	7.370
Absolute Deviation	:	0.2067
Standard Deviation	:	0.2938
Variance	:	0.8633E-01
Skewness	:	-2.193
Kurtosis	:	15.07
Minimum	:	4.697
2.5% percentile	:	6.751
25% percentile	:	7.224
Median	:	7.387
75% percentile	:	7.551
97.5% percentile	:	7.827
Maximum	:	8.012
Number of Iterations	:	500

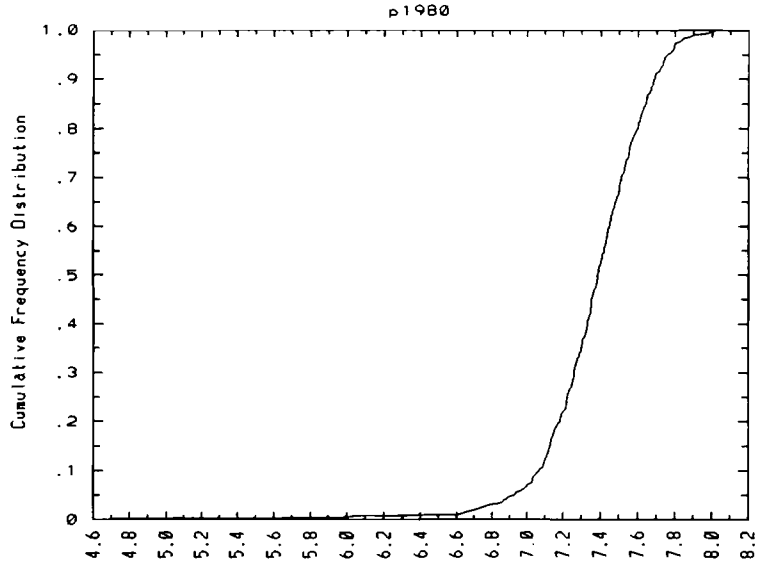

Figure III.7 Cumulative distribution of RAINS Lake Module pH response in 1980 in an insensitive sub-region with deposition of 5 g/m2/yr.

Mean	.	7.181
Absolute Deviation	.	0.3573
Standard Deviation	.	0.6015
Variance	.	0.3618
Skewness	.	-3.019
Kurtosis	.	10.56
Minimum	.	3.975
2.5% percentile	.	4.889
25% percentile	.	7.097
Median	.	7.318
75% percentile	.	7.496
97.5% percentile	.	7.765
Maximum	.	7.932
Number of Iterations	.	500

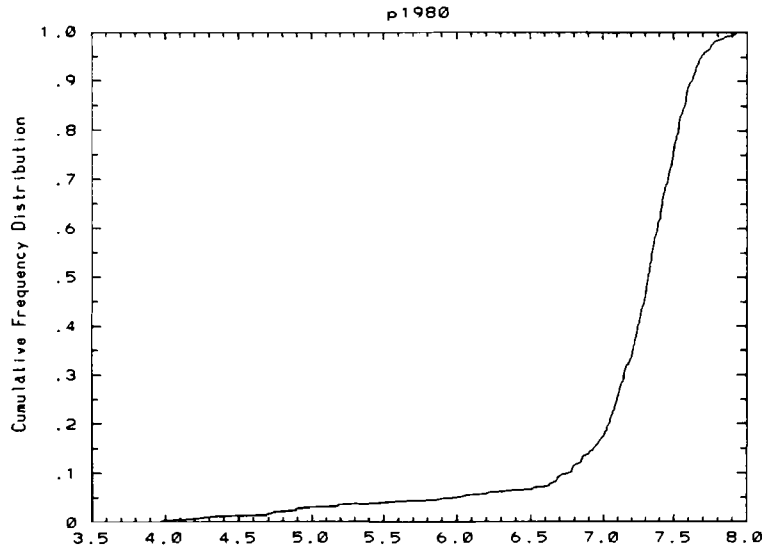

Figure III.8 Cumulative distribution of RAINS Lake Module pH response in 1980 in an insensitive sub-region with deposition of 10 g/m2/yr.