ACIDIFICATION IN EUROPE: A SIMULATION MODEL FOR EVALUATING CONTROL STRATEGIES

J. Alcamo, M. Amann, J.-P. Hettelingh, M. Holmberg,
L. Hordijk, J. Kämäri, L. Kauppi, P. Kauppi,
G. Kornai and A. Mäkelä,
International Institute for Applied Systems Analysis
Laxenburg, Austria

RR–88–2
January 1988

Reprinted from *AMBIO*, volume 16, No. 5 (1987).

INTERNATIONAL INSTITUTE FOR APPLIED SYSTEMS ANALYSIS
Laxenburg, Austria

Research Reports, which record research conducted at IIASA, are independently reviewed before publication. However, the views and opinions they express are not necessarily those of the Institute or the National Member Organizations that support it.

Reprinted with permission from *AMBIO*, **16**(5), 1987.

Printed by Bohusläningens Boktryckeri AB, Uddevalla, Sweden and Novographic, Vienna, Austria

FOREWORD

One of IIASA's principal goals is to narrow the gulf between scientists and decision makers. The transboundary flow of air pollutants and the resulting acidification of the environment is one problem which needs clear decisions based upon the best available scientific information.

Since late 1983 the Acid Rain Project has been using techniques of systems analysis to develop an integrated model which simulates the transboundary flow of acidifying air pollution from the emission sources to the environment. Given a certain pattern of energy use, the model can estimate geographical patterns of sulphur deposition, its effects upon certain aspects of the environment, and the costs of any abatement measures that are applied. Alternatively the model can indicate the optimum geographical distribution of emission reductions, given an environmental target.

This article is a status report on the decision making tool that has been developed.

R.W. SHAW
Leader
Acid Rain Project

Acidification in Europe: A Simulation Model for Evaluating Control Strategies

Report

By Joseph Alcamo, Markus Amann, Jean-Paul Hettelingh, Maria Holmberg, Leen Hordijk, Juha Kämäri, Lea Kauppi, Pekka Kauppi, Gabor Kornai and Annikki Mäkelä

RAINS (*Regional Acidification INformation and Simulation*) is an integrated model of acidification in Europe designed as a tool for evaluating control strategies. It is currently sulfur-based, but is being expanded to include nitrogen species. Emphasis of the model is on the transboundary aspects of the acidification problem. Model computations are performed on a personal computer. Linked submodels are available for SO_2 emissions, costs of control strategies, atmospheric transport of sulfur, forest soil and groundwater acidity, lake acidification, and the direct impact of SO_2 on forests. The model can be used for scenario analysis, where the user prescribes a control strategy and then examines the cost and environmental consequences of this strategy, or for optimization analysis, in which the user sets cost and deposition goals, and identifies an "optimal" sulfur-reduction strategy. Preliminary use of the model has pointed to 1. the importance of examining long-term environmental consequences of control strategies, and 2. the cost advantages of a cooperative European sulfur-reduction program.

INTRODUCTION

There is an information gap between scientists who study acidification in Europe and those responsible for taking action on the problem. The consequences of this gap were summarized at a recent meeting on "environmental diplomacy", where it was claimed that governments were left unsure of causes and effects regarding environmental matters, and were unable to balance short-term costs with long-term benefits (1).

This communication cannot be improved simply by gathering additional data, because sensible control strategies must be based on understanding the entire acidification system. We can certainly measure the individual parts of Europe's acidification system, for instance, by monitoring sulfur dioxide emissions from certain power plants; tracking the force and direction of the wind; or measuring air pollutant concentrations and acidity levels of lakes and soil in remote areas. But to see how these different parts interact with one another takes either fantastic imagination—or a method for synthesizing this information, as in a mathematical model. The RAINS (*Regional Acidification INformation and Simulation*) model of Europe, developed at the International Institute for Applied Systems Analysis (IIASA), describes this system and provides information useful for both policy advisors interested in control strategies and scientists interested in a comprehensive view of the problem. An earlier version of this model is presented elsewhere (2, 3).

The emphasis of the model is on the transboundary aspect of air pollution in Europe with the principal aim to present a spatial and temporal overview of the problem. Hence, the spatial coverage of RAINS is all of Europe, including the European part of the USSR, and the time horizon begins in 1960 to permit checking of historical calculations and extends to 2040 to allow examination of long-term consequences of control strategies.

The model is currently sulfur-based because of the principal role of sulfur as a precursor of acid deposition. However, the model is being expanded to include nitrogen emissions, transport, deposition and impacts.

The model deals with pollution generation, atmospheric processes, and environmental impacts; each of these subjects is described by submodels which are connected as shown in Figure 1.

The design of each submodel is influenced by the broad spatial/temporal character of the model. Each submodel is as simple as possible, to facilitate interactive use and comprehension, but maintains enough description to capture the essential dynamics of the acidification system. This will be explained as each submodel is described. Because of the large spatial coverage and long time horizon, the time step of calculations must be rather large (a season or a year) and the spatial aggregation must also be large (150×150 km for deposition on 0.5° latitude × 1.0° longitude for the environmental impact submodels).

The development of RAINS is among the first efforts to build an integrated model covering regional- or interregional-scale air pollution problems. Elsewhere in Europe (4, 5) and in North America (6, 7) models for the evaluation of transboundary air pollution are being built. There are also government sponsored integrated modeling studies underway in Finland, The German Democratic Republic, The Netherlands, and Norway (8).

In this article we describe each of the submodels which make up RAINS, as well as how they are linked, how the RAINS model is tested and used, and some tentative conclusions from our study thus far.

ENERGY USE AND POLLUTANT EMISSION

SO_2 emissions in Europe originate from a variety of anthropogenic and natural sources. Many investigators have noted that the amount of sulfur on an annual basis from volcanoes, marshes, and other natural origins is insignificant in Europe compared to anthropogenic emissions (9, 10). Consequently, in the RAINS model we concentrate on anthropogenic sources and compute sulfur emissions for each of several emission-producing economic sectors in each of 27 European countries. Sulfur emissions are calculated by mass balance which accounts for the energy consumed in each sector together with fuel characteristics such as sulfur content, heat value, and amount of sulfur retained by combustion (Box 1). Emissions from different sectors, plus sulfur emissions from industrial processes are summed to obtain country emissions.

Energy data for 1960–80 are taken from UN statistics provided by the Economic Commission for Europe (11). Because of the great uncertainty in future energy use, the model user is given the option of selecting one of three energy pathways. These include an *"official" energy pathway*, a *maximum natural gas utilization energy pathway*, and a *nuclear phase-out pathway*.

The second and third pathways are based on recent IIASA studies of future energy use in Europe. The pathway for *maximum natural gas utilization*, as the name implies, investigates the possibilities of increased introduction of natural gas to Europe (12).

Box 1: SO_2—Emissions submodel

Symbols:

E	energy use
hv	heat value
sc	sulfur content
sr	fraction of emissions retained in ash
x	fraction of emissions removed by pollution control
S	sulfur emissions
S^P	sulfur emissions from industrial (non combustion) processes
i	country
j	fuel type
k	economic sector
l	abatement technology
t	time

Sulfur emissions calculation:

Sectoral emissions per fuel:

$$S_{i,j,k}(t) = \sum_l E_{i,j,k,l}(t) \frac{sc_{i,j,k}}{hv_{i,j}} (1-sr_{j,k}) (1-x_{i,k,l})$$

Total sulfur emissions per country:

$$S_i(t) = \sum_j \sum_k S_{i,j,k}(t) + S_i^P(t)$$

The *nuclear phase-out pathway* is based on the assumption that no nuclear-power plants are built after 1990 in Western Europe and the existing ones are used for their planned life of 25 years (13).

The *"official" energy pathway* consists of official government projections as compiled by the International Energy Agency (14) for Western Europe and the Economic Commission for Europe (15) for Eastern Europe and the USSR. The Western European projections imply that the use of coal and nuclear power will increase substantially. Authors of the report point out, however, that "national projections may reflect policy goals and are not necessarily 'most likely case' forecasts". Of course this comment can be applied to each pathway. For this reason the user of RAINS has the option to interactively input their own energy projections for one or several countries. The user can experiment with drastically changed fuel mixes to investigate their effect on emissions and environmental impacts. However, RAINS performs a consistency check on these user-prescribed pathways in that final energy demand is matched with energy supply. Moreover, RAINS produces a warning signal whenever a user makes an unreasonable assumption (e.g. by assuming a very high hydropower capacity in a country with limited streamwater resources).

The submodel accounts for five emission-producing sectors: *conversion* (e.g. refineries), *power plants, domestic, industry* and *transportation*. Eight fuels may be used in each sector: *brown coal, hard coal, derived coal* (e.g. brown coal briquettes and coke), *light oil* (e.g. gasoline), *medium destillate* (gas oil), *heavy oil, gas*, and *"other fuels"*. The gas and "other fuels" sectors are assumed to produce no sulfur emissions.

Considering the aggregated nature of the sectors and uncertainty of inputs, a pragmatic approach is taken to calibrate model parameters. Parameters are calibrated to 1980 SO_2 emissions from each country (16) because this is the most complete and internationally consistent data set currently available. Calibration takes into account many data derived from international statistics on fuel, fuel trade, and sulfur content of fuels.

Nitrogen oxide emissions cannot be calculated in the same way as sulfur emissions because they originate not only from nitrogen in fuel, but also from nitrogen in air. These two components are termed *fuel* NO_x and *thermal* NO_x, respectively. Fuel NO_x can be calculated by performing a mass balance as in the sulfur-emission calculations, but thermal NO_x strongly depends on combustion characteristics which require a detailed description of NO_x-producing sectors for each European country. As an alternative, we have derived sector and fuel-specific NO_x emission factors from a regression of emissions on fuel use per sector which are applicable to many countries (17). By using these factors we come close to the official NO_x emissions of many Western European countries. For Eastern European countries, however, more assumptions will be needed to take into account differences between Eastern and Western European energy systems.

POLLUTION CONTROL AND COST ANALYSIS

There are basically four ways to reduce sulfur emissions originating from energy combustion: 1. energy conservation, 2. fuel substitution, 3. use of low sulfur fuels and 4. desulfurization during or after fuel combustion. For options 2 to 4 RAINS contains a formal procedure to estimate potential reductions and costs of their application. Costs of energy conservation strategies are not investigated within RAINS, because goals other than pollution control may motivate energy conservation policies.

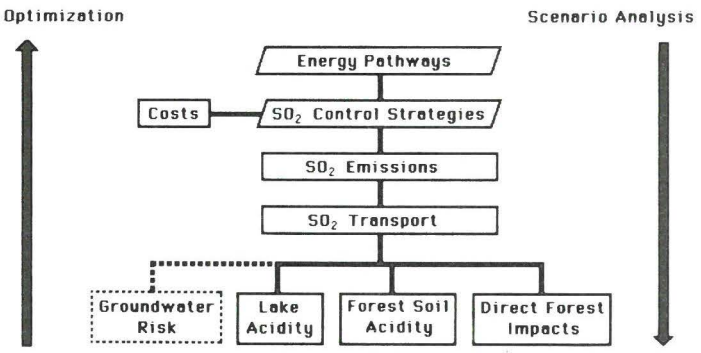


Figure 1. A schematic overview of the RAINS model.

Table 1. Pollution control options.

		Low Sulfur	Combustion modification retro	Combustion modification new	Flue gas desulfurization retro	Flue gas desulfurization new	Regeneration process
Conversion	Hard coal				X		
	Heavy fuel oil				X		X
Powerplants	Brown coal		X	X	X	X	
	Hard coal	X	X	X	X	X	
	Heavy fuel oil	X			X	X	
Domestic	Hard coal	X					
	Coke, Briquettes	X					
	Gasoil	X					
	Heavy fuel oil	X					
Transport	Gasoil	X					
Industry	Hard coal	X	X		X		X
	Coke				X		X
	Gasoil	X					
	Heavy fuel oil	X			X		X

Box 2: Cost analysis submodel

Symbols:

I_{an}	annualized investment costs
OM_{fix}	fixed operation and maintenance costs
OM_{var}	variable operation and maintenance costs
xp	efficiency of process emissions removal
cp	unit costs for process emissions removal
c_l	unit costs for direct abatement
cf	price differential for fuel substitution
E^*	energy use in original scenario
S^{*P}	unabated process emissions

Country specific data:

sc	sulfur content
hv	heat value
sr	sulfur retained in ash
bs	average boiler size
pf	capacity utilization
q	real interest rate
c^e, c^l, c^s, c^d	prices for electricity, labor, sorbents and waste disposal

Technology specific data:

I	investment function
v	relative flue gas volume
lt	life time of plant
x	sulfur removal efficiency
α	ratio sulfur/sorbents
f_l	maintenance costs
λ^e, λ^l, λ^s, λ^d	specific demand for energy, labor, sorbents and waste disposal

Direct abatement costs, C^d:

(for reasons of simplicity indices for countries (i), fuels (j) and sectors (k) are omitted where possible)

- pollution control measures without investments: c_l are taken from the literature
- abatement technologies, which require investments:

$$I_{an} = f\,(I,\ bs,\ v_j,\ lt,\ q)$$

$$OM_{fix} = f\,(I,\ bs,\ v_j,\ f_l)$$

$$OM_{var} = f\,(\alpha,\ c^e,\ \lambda^e,\ c^l,\ \lambda^l,\ c^s,\ \lambda^s,\ c^d,\ \lambda^d)$$

$$c_l = \frac{I_{an} + OM_{fix}}{pf} + OM_{var}\,\frac{sc}{hv}\,(1-sr)\,x$$

$$C^d = \sum_j \sum_k \sum_l E\ c_l$$

Fuel switching costs, C^f:

$$C^f = \sum_j \sum_k E\ cf - \sum_j \sum_k E^*\ cf$$

Control costs for process emissions, C^P:

$$C^P = S^{*P}\ xp\ cp$$

Total pollution control costs:

$$C_{i,\,t} = C^d + C^f + C^P$$

Fuel Substitution

Fuel substitution can be performed within ranges which are derived from the differences between the energy pathways. Consistency of the energy balance is preserved, taking into account the different combustion efficiencies of fuels. The cost calculation submodel provides rough cost estimates for fuel substitution policies by using country-specific price differentials between fuels.

Low Sulfur Fuels

The costs of low sulfur fuels are derived from observations of the world market prices for hard coal and from an analysis of international cost data for fuel desulfurization of oil products.

Desulfurization

We describe desulfurization during or after the fuel combustion by three technologies, each having different costs and efficiencies. These technologies are combustion modification, flue gas desulfurization (FGD) and regenerative processes. Whereas combustion modification requires only a few additional investments

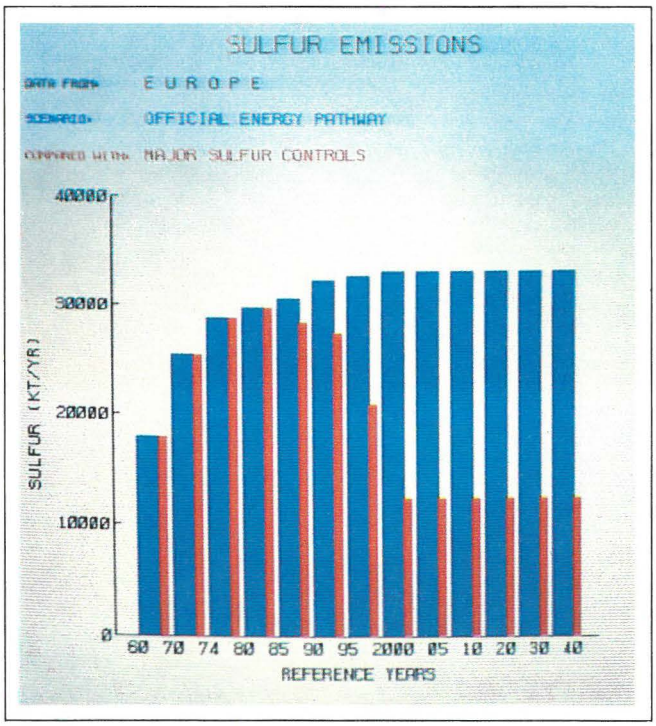


Figure 2. **SO_2 emissions for Europe for two scenarios.** ***Official Energy Pathway—No Controls* and *Major Sulfur Controls.***

(resulting in moderate sulfur removal efficiencies), flue gas desulfurization (e.g. wet limestone scrubbing) usually results in both higher investment costs and cleaning efficiencies. For extreme sulfur-reduction scenarios costs are based on the most efficient regenerative flue gas desulfurization process (i.e. the Wellman-Lord process). Of course, not all options are applicable to all economic sectors or fuels. Table 1 gives an overview of the sector/technology combinations considered in RAINS. Since in most countries thermal power plants have the biggest share of the sulfur emissions, a distinction is made between old installations, to which retrofit technologies can be applied and—depending on the selected energy pathway—new plants, where pollution control can be achieved at lower costs.

The costs for implementing emission reductions are derived by the procedures shown in Box 2, which take into account country and sector-specific parameters (18). The resulting abatement cost coefficients incorporate the most important cost influencing circumstances of the European countries in an internationally comparable way. Since the emphasis of RAINS is on the transboundary aspect of air pollution, the objective of the cost submodel is not to provide exact cost estimates, but to create a common basis for international cost comparisons.

To use the cost submodel of RAINS one first has to select an existing energy pathway or create a new pathway. SO_2 emission control strategies can then be specified in three different modes. In the first mode a user can create a control strategy by applying combinations of three emission-reduction methods: fuel substitution, the use of low sulfur fuels, and desulfurization. After the user has specified the amounts of energy per sector and fuel to which each of these methods has to be applied, RAINS provides both the related costs and the achieved SO_2 emission reductions. In the second mode a user specifies amounts of emission reductions per country, and RAINS estimates the optimal abatement costs to achieve these reductions, using its country-specific cost functions. The third mode is used to compute an international cost optimum for reducing deposition to a specified level.

The results of a sample control strategy, *Major Sulfur Controls*, on SO_2 emissions are shown in Figure 2, which at the same time provides an example of computer screen output. This scenario is based on the *Official Energy Pathway*, with controls applied as listed in Box 3. For comparison we present the *Official Energy Pathway* without controls. In Table 2 we present a *30% Reduction All Europe* scenario, which assumes that every European country will accomplish a 30 percent reduction of SO_2 emissions relative to their 1980 levels. This reasonable extension of current policy will be used as a reference case for the remainder of the paper. In Table 2 SO_2 emission levels are given for the year 1980, for the three scenarios described here and for a *Deposition Limit* scenario described later.

Uncontrolled emissions from Europe in 1980 are estimated to be 29.8 MT $\cdot$ yr^{-1} (measured as sulfur). Emissions in the year 2000 for the *Major Sulfur Controls* scenario are estimated to decrease to 12.5 MT $\cdot$ yr^{-1}, which is close to their 1940s level (19, 20), and substantially lower than emissions from the *30% Reduction* scenario. Emissions for both scenarios are assumed to level off after the year 2000.

Box 3: Scenario overview

a) *Official Energy Pathway*
As published by IEA and ECE; no pollution control assumed.

b) *30% Reduction All Europe*
Based on the Official Energy Pathway,
SO_2 emissions are reduced by 30% based on the 1980 level.

c) *Major Sulfur Control (MSC)*
As an example of a user specified emission reduction strategy the MSC-Scenario implements in all countries (based on the Official Energy Pathway) strong pollution control in the following way (shown for the year 2000; the policy is assumed to be phased in from 1985 onwards):

Sector	Control option	Share of energy treated	SO_2 removal efficiency	Resulting sectoral SO_2 removal
Conversion	FGD	0.90	0.90	0.81
Powerplants	FGD	0.90	0.90	0.81
Industry	FGD	0.50	0.90	0.45
Domestic	low S	1.00	0.50	0.50
Transport	low S	1.00	0.50	0.50

d) *Deposition Limit*
A cost optimal solution for reducing the maximum deposition level within Europe to 5 g $\cdot$ m^{-2} $\cdot$ yr^{-1}.

Table 2. SO_2 emissions (kilotons sulfur).

Country	1980	30% Reduction	Deposition Limit 5 g $\cdot$ m^{-2} $\cdot$ yr^{-1} (2000)	Major Sulfur Controls (2000)
Albania	39	27	39	15
Austria	159	111	33	89
Belgium	432	303	73	142
Bulgaria	508	355	508	363
Czechoslovakia	1832	1282	384	592
Denmark	226	158	226	77
Finland	294	206	263	100
France	1657	1160	180	448
FRG	1602	1121	264	464
GDR	2415	1691	640	996
Greece	345	242	345	226
Hungary	813	569	600	352
Ireland	119	83	119	71
Italy	1898	1328	1172	640
Luxembourg	20	14	3	12
Netherlands	243	170	24	155
Norway	72	51	72	43
Poland	1741	1219	636	841
Portugal	130	91	130	91
Romania	757	530	757	566
Spain	1879	1315	1879	966
Sweden	243	170	192	100
Switzerland	67	47	27	38
Turkey	497	348	497	779
UK	2342	1639	658	967
USSR	8588	6012	1822	2878
Yugoslavia	837	586	837	446
Europe	29 752	20 826	12 407	12 455
% Reduction	–	30	58	58

ATMOSPHERIC TRANSPORT

Since the typical residence time of SO_2 in the atmosphere is in the order of one to two days (21), SO_2 emissions from one European country are often deposited in another country. Dry and wet removal processes control the atmospheric residence time of sulfur. Model calculations indicate that their relative contribution to total deposition varies throughout Europe; dry deposition is more important close to the high densities of sulfur emissions and wet deposition in more remote areas (22). These removal processes, together with meteorologic transport, are simulated by long-range transport models.

In RAINS the atmospheric transport submodel computes SO_2 air concentration

and sulfur deposition in Europe due to the sulfur emissions in each country, and then sums the contributions from each country with a background contribution to compute the total sulfur deposition or SO_2 concentration at any grid location (Box 4). The submodel consists of a transfer matrix based on a Lagrangian model of long-range transport of air pollutants in Europe, developed under the Organization of Economic Cooperation and Development (OECD) and later under the Cooperative Program for the Monitoring and Evaluation of Long Range Transmission of Air Pollutants in Europe (EMEP). This model accounts for the effects of winds, precipitation, and other meteorological and chemical variables on sulfur deposition and air concentration and has been extensively tested against observations (23). The EMEP model computes SO_2 and SO_4^{2-} air concentrations along wind trajectories throughout Europe.

Use of a transfer matrix in this fashion assumes that there is a linear relationship between SO_2 emissions and computed sulfur deposition or SO_2 concentration. Model experiments conducted with the EMEP model found this to be a reasonable assumption for a time scale of one year and for the relationship between country emissions and grid deposition (24).

Model calculations in this paper are based on a transfer matrix made available to IIASA by the EMEP Meteorological Synthesizing Center-West in Oslo, Norway. A new version of this model now exists (25) but was not implemented in RAINS at the time of this publication. Transfer matrices based on the EMEP model, but for other years can be used as well. Matrices from other long-range transport models will also be used a soon as they become available.

In Figure 3 we present several examples of total sulfur deposition output from the sulfur transport submodel. Figure 3a depicts the 1980 situation in which most of Central Europe, as well as parts of the United Kingdom and USSR receive more than 4 $g \cdot m^{-2} \cdot yr^{-1}$ deposition. Only the northernmost and southernmost areas of Europe, which are most distant from concentrated emissions, receive less than 1 $g \cdot m^{-2} \cdot yr^{-1}$. Figure 3b presents an extreme (but unrealistic) case in which no pollution controls are implemented in any European country in the year 2000. The area covered by more than 4 $g \cdot m^{-2} \cdot yr^{-1}$ spreads to southeastern Europe. For the case of a European-wide 30 percent reduction in sulfur emissions (Figure 3c), most of Europe receives deposition between 1 and 4 $g \cdot m^{-2} \cdot yr^{-1}$, and the area covered by greater than 4 $g \cdot m^{-2} \cdot yr^{-1}$ diminishes to a smaller part of Central Europe. Further improvement is seen in the *Major Sulfur Controls* scenario for the year 2000 (Figure 3d). The area with deposition less than 1 $g \cdot m^{-2} \cdot yr^{-1}$ greatly increases and includes virtually the entire Nordic area. Finally Figure 3e shows the results of an optimization scenario explained later in the text in which maximum deposition throughout Europe is held at 5 $g \cdot m^{-2} \cdot yr^{-1}$. (Some small parts of Europe have greater than 5 $g \cdot m^{-2} \cdot yr^{-1}$ deposition because of upper limits placed on SO_2 emission reductions in the optimization analysis.)

While it may be acceptable to linearly approximate sulfur source-receptor relationships, it is more difficult to do so for NO_x long-range transport because of the more complicated atmospheric chemistry involved. Nevertheless, some NO_x long-range transport models with rudimentary chemistry are beginning to show promising results when compared to observations over large time and space scales. Our strategy will be to include results of these models in RAINS as transfer matrices with correction factors to account for non-linear chemistry.

In another development, we are implementing transfer matrices based on a long-range transport model of ammonia (26) and NO_x (27) in Europe. Our ultimate aim is to combine output from NH_4-N and NO_x-N to estimate total nitrogen deposition at various locations in Europe.

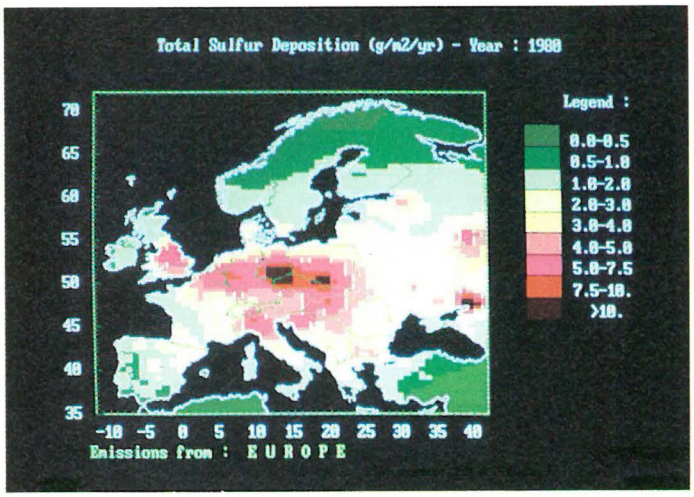
SOIL ACIDIFICATION

Soil acidification is an important link between air pollution and damage to the terrestrial and aquatic environment. The ability of soil to buffer acid deposition is a key factor in regulating the long-term surface water and groundwater acidification. Soil acidification has also been related to forest dieback via its effect in the tree root zone (28).

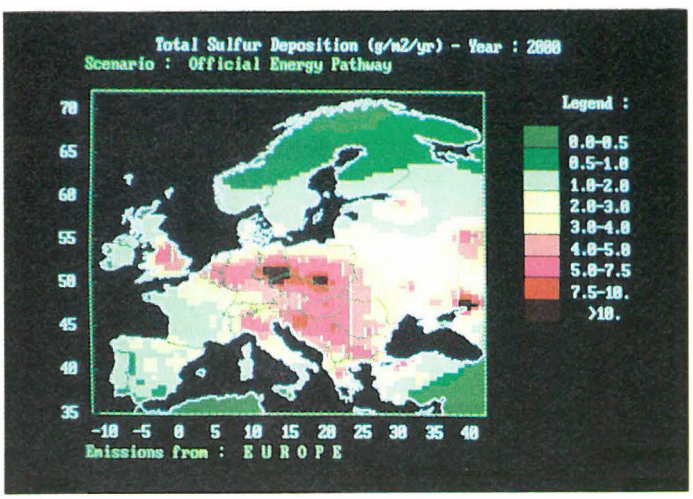
Soil acidification has been defined as the decrease in acid neutralization capacity of

Figure 3. Maps of total sulfur deposition (a) 1980 (b) *Official Energy Pathway*, 2000 (c) *30% Reduction*, 2000 (d) *Major Sulfur Controls*, 2000 (e) 5 $g \cdot m^{-2} \cdot yr^{-1}$ *Deposition Limit*, 2000.

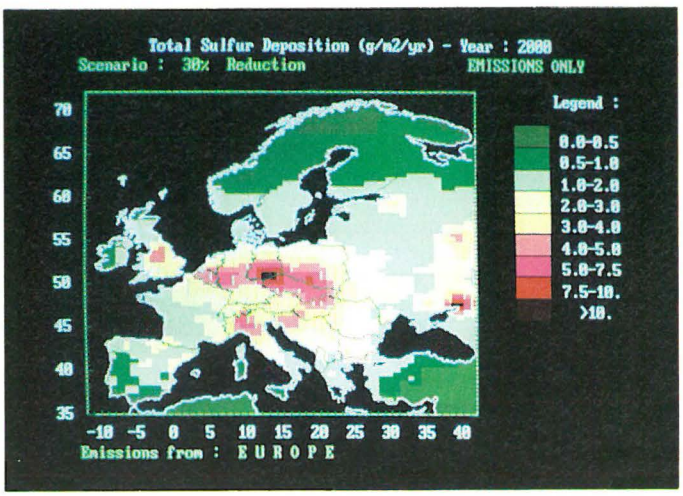
(a)



(b)



(c)



the soil (29). Weathering of base cations is the process in mineral soils that generates neutralizing capacity. Therefore, soil acidification proceeds when the rate of acid input exceeds the weathering rate. Various phenomena are associated with acidification: pH decline, decrease in base saturation, increase in soluble aluminum concentrations, and a general increase in ion fluxes through soil.

The RAINS soil submodel focuses on year-to-year development of forest-soil acidification in an idealized 50 cm deep soil layer. Soil acidity in this layer is computed from *acid load* and buffering characteristics of the soil. Acid load (the flux of protons to soils) is calculated by assuming that all sulfur deposition is oxidized. Buffering characteristics are divided into "buffer capacity," the total reservoir of buffering compounds in soil, and "buffer rate," the maximum potential rate of the reaction between buffering compounds and acid load. In some cases where buffer capacity is high, a low buffer rate may nevertheless limit the ability of soil to buffer the acid load. Both characteristics reflect intrinsic properties of soil such as lime content, silicate weathering rate, cation-exchange capacity, and base saturation.

To compute soil acidity, the model compares the cumulative load to the buffer capacity, and the rate of acid loading on a year-to-year basis with the buffer rate (Box 5). Depending upon the acid load there is either a recovery or an exhaustion of the prevailing cation-exchange capacity. In case the deposition rate of strong acids is lower than the silicate buffer rate, the weathering gradually fills up the cation-exchange complex and the model computes a recovery. The hydrogen ion concentration is calculated either on the basis of base saturation, i.e. the fraction of cation exchange sites occupied by base cations, or according to equilibrium with solid phases of aluminum. Initialization of the model variables was based on chemistry information available on European soils, and on the soil thickness selected to approximate the tree root zone (30).

A sensitivity analysis of the soil model has shown that the model is particularly sensitive to base saturation, silicate buffer rate, and a "filtering factor" discussed later in this article (31). Base saturation needs special attention because of the large uncertainty of its initial value. The model is sensitive to changes in the silicate buffer rate only if this rate is of the same magnitude as the acid load, as in areas distant from pollutant sources. However, if deposition decreases in the future, then the silicate buffer rate will become important in larger areas. In general, the sensitivity tests pointed out the importance of initial conditions of the soil.

Figure 4 presents an example of soil model output in the form of a map of the country-by-country status of soil acidity in the year 2000 resulting from the *30% Reduction* and *Major Sulfur Controls* scenarios. This figure combines information about computed acidity levels of different soils with a data base of forest coverage throughout Europe. The pH levels and the year can be chosen by a model user. In our example we have selected values of 4.0 and 4.3, values which reflect a doubling of hydrogen-ion concentration. The figure demonstrates that in Central Europe forest soils are frequently in the low pH classes. In Southern Europe, Scandinavia and the USSR the highest pH class dominates. The *Major Sulfur Controls* scenario results in higher pH levels in Central Europe compared to the *30% Reduction* scenario. Differences between the two scenarios taper off with

Box 4: Sulfur transport submodel

Symbols:

a	transfer coefficient; deposition per unit emissions	$k_{w,\,c_1}$	wet removal rate for SO_2
		$k_{w,\,c_2}$	wet removal rate for SO_4^{2-}
		n	grid element
b	background deposition	Q	emission flux
c_1	SO_2 air concentration	S_i	emissions from country i
c_2	SO_4^{2-} air concentration	t	time
$\frac{D}{dt}$	total time derivative	v_d	dry deposition velocity for SO_2
		w_d	dry deposition velocity for SO_4^{2-}
h	mixing height	α	local deposition coefficient
i	country	β	coefficient accounting for SO_4^{2-} emissions
k_t	transformation rate		

To compute deposition in RAINS:

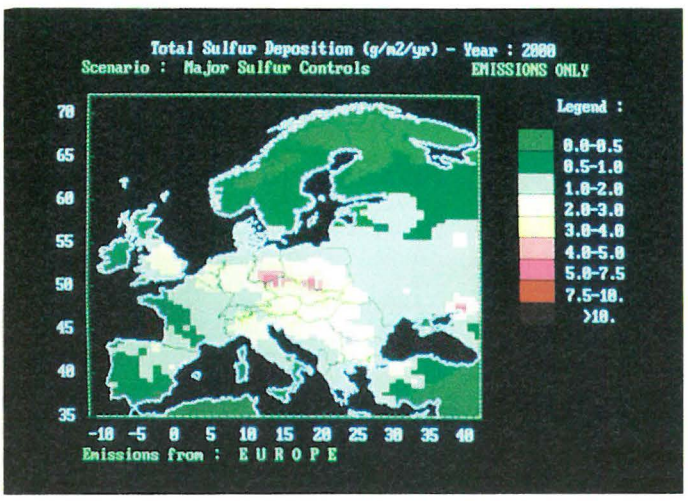
$$d_n(t) = \sum_i S_i(t)\, a_{i,n} + b_n$$

The transfer coefficients, $a_{i,n}$ are derived from the EMEP model of sulfur transport in Europe with the basic equations:

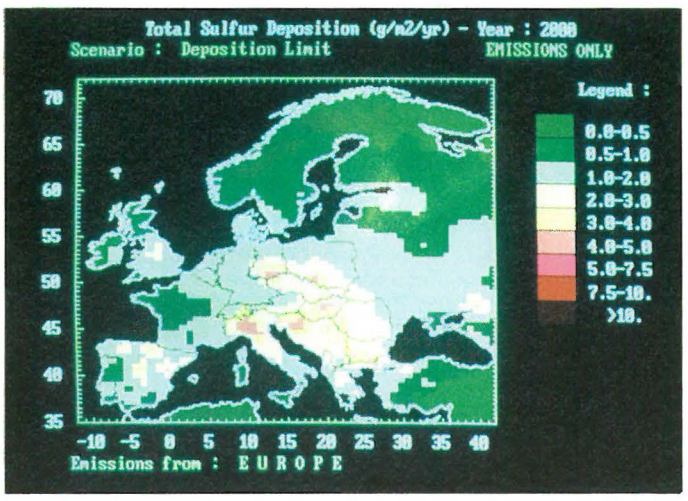
$$\frac{Dc_1}{dt} = -\left[\frac{v_d}{h} + k_t + k_{w,c_1}\right] c_1 + (1-\alpha-\beta)\,\frac{Q}{h}$$

$$\frac{Dc_2}{dt} = -\left[\frac{w_d}{h} + k_{w,c_2}\, c_2\right] + k_t c_1 + \beta\,\frac{Q}{h}$$

(d)



(e)



Box 5:
Soil acidification submodel

Symbols:

BC_{CE}	prevailing cation-exchange capacity
CEC_{tot}	total cation-exchange capacity
ac	acid load rate to the soil
wr	silicate weathering rate
c_H	hydrogen ion concentration
c_{Al}	aluminum ion concentration
K_{so}	equilibrium constant for aluminum solubility
t	time

Weathering and cation exchange:

$$BC_{CE}(t) = BC_{CE}(t-1) - (ac(t) - wr)$$

Equilibrium concentrations:

$$c_H(t) = f(BC_{CE}(t),\ CEC_{tot})$$

$$c_{Al}(t) = K_{so}c_H^3(t)$$

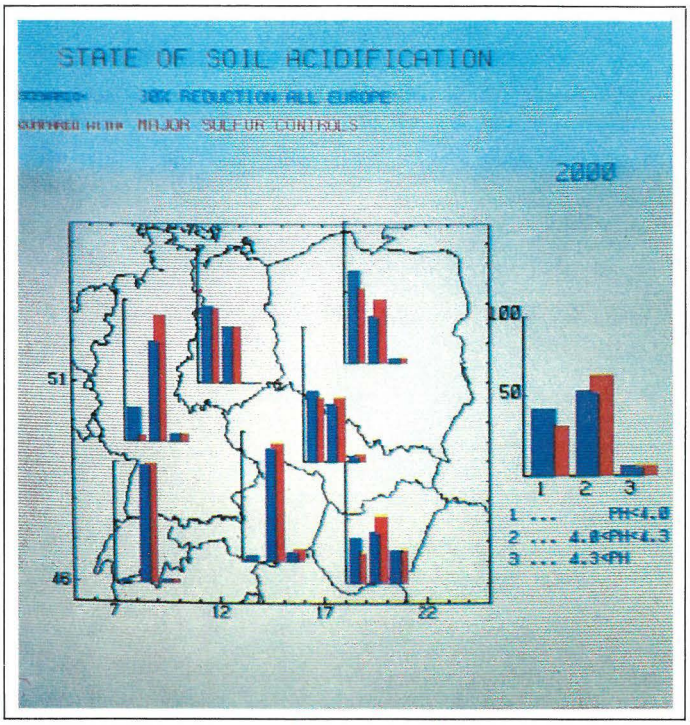


Figure 4. Distribution of Central European forest soils in pH classes for *30% Reduction* (left bars) and *Major Sulfur Controls* scenarios in the year 2000. The bar chart at the right hand side of the picture gives the aggregated distribution for all countries shown.

distance from Central Europe. This is consistent with the smaller differences in deposition between the two scenarios in these areas (Figures 3c and 3d).

Seen over a longer time horizon the *Major Sulfur Controls* scenario shows an even greater improvement over the *30% Reduction* scenario (Figure 5). For example, about 5 percent of forest soils in the Federal Republic of Germany are in the lowest pH class for the first scenario in the year 2000 compared to 27 percent for the latter scenario. For the year 2040 the area of forest soils in the lowest pH range has only slightly increased for the *Major Sulfur Controls* scenario (6 percent), whereas it has nearly doubled for the *30% Reduction* scenario (53 percent). This pattern is similar for many other Central European countries. It is also worth noting that forest-soil acidification continues to increase in Central Europe for the *Major Sulfur Controls* scenario in which total European emissions are reduced by nearly 60 percent from 1980. This increase is small but noticeable as illustrated by Figure 6, which presents time histories of areas in Central Europe where forest soil pH is less than or equal to 4.0. Note the continuing large increase in area with low pH under the *30% Reduction* scenario.

LAKE ACIDIFICATION

Lake acidification is a well-documented problem in many mountainous and forested regions of Europe. In Sweden, for example, it has been estimated that acidification affects 15 000 of 85 000 lakes larger than one hectare in area and seriously affects 1800 of these (32). There is strong evidence that lake acidification can result from acidic runoff that is inadequately buffered by soils in the lake's catchment. The extent of lake acidification also depends on the amount of snowmelt, flow paths of runoff, lake chemistry, and other physical and chemical processes. The RAINS lake submodel attempts to provide a quantitative overview of the key processes (Box 6).

A simple two-layer structure is used for simulating the routing of internal flows (33). The terrestrial catchment is segmented into snowpack and two soil layers (A- and B-reservoirs). Precipitation is routed into quickflow, baseflow, and percolation between soil layers. Physically, the flow from the upper reservoir can be thought of as quickflow, which drains down the hillsides as piped flow or fast throughflow and enters the brooks directly. This water is mainly in contact with humus and the upper mineral layer. The B-reservoir in the model provides the baseflow, which presumably comes from deeper (>0.5 m) soil layers.

To compute the ion concentrations of the internal flows, the same analytical approach is applied as in the RAINS soil acidification model (Box 5). The contribution of the soil reservoir to the alkalinity of the surface water is assumed to equal the amount of weathered base cations minus the acid load. The leaching of acidity to surface waters is simulated on the basis of simulated concentrations in the soil solution and the discharges from both reservoirs.

The change in lake water chemistry is predicted by means of equilibrium expressions given for inorganic carbon species. The carbonate alkalinity is assumed to be the only significant buffering agent. It originates from both the terrestrial catchment and from in-lake processes (34). The ion loads to the lake are mixed within a layer which depends on location and season. In practice, meteorological and hydrological variables are summed over the whole year and simulations are carried out using an annual time step. The risk for aquatic impacts is estimated on the basis of simple threshold pH and alkalinity values. These characteristics are most likely to indicate damage to fish populations and other aquatic organisms.

The approach for assessing regional lake water impacts has two distinct levels. At the first level the catchment model is able to analyze changes over time in the chemistry of any specific lake. At the second level, the catchment model is regionalized by expanding the set of parameters to include characteristics of a large number of lakes within a particular region. To regionalize the model, a Monte-Carlo method is used to select combinations of input parameters that produce the distribution of output variables observed in the study region (35). A subset of parameter combinations that produce the actual observed present-day lake acidity distribution in each lake region is obtained. Assuming that the set of input values obtained in this filtering procedure is representative for real catchments in the study region, this ensemble can be used for the scenario analysis of the response of lake systems to different patterns of acidic deposition. As a result this method for scenario analysis produces frequency distributions for lake pH and alkalinity for any scenario and year.

Differential sensitivity of model output has been calculated by a Monte-Carlo method in which variance of all parameters has been set to one percent of their nominal value. The sensitivity analysis shows that catchment soil thickness explains over 35 percent of the variability in computed 1980 lake pH levels. Initial base

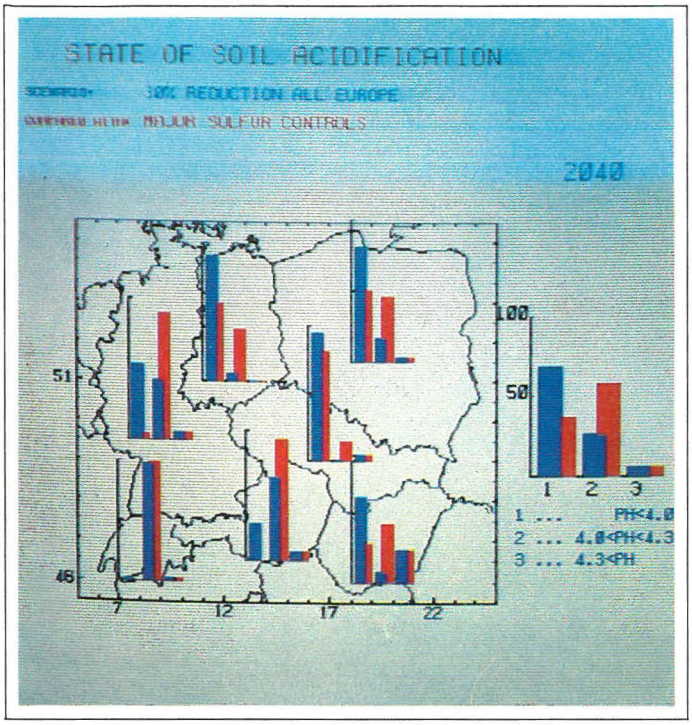


Figure 5. As Figure 4, in the year 2040.

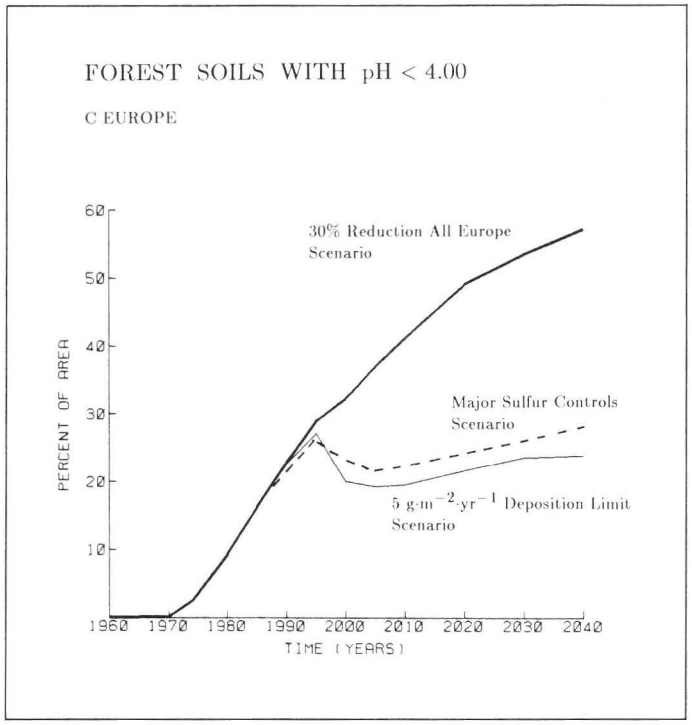


Figure 6. Percentage of Central European forest soils with pH less than 4.0 for the scenarios. Total geographic area considered is represented by the rectangle in Figures 4 and 5.

saturation in the B soil layer and silicate buffer rate, explain 15 percent and 14 percent, respectively, of the total lake pH variability. Melting rate, evapotranspiration rate, field capacity and the hydraulic conductivity at saturation level each explain less than 10 percent of the variability of results. The remaining 38 parameters are relatively unimportant, affecting the model output by less than one percent.

The calibrated soil thickness has a moderately large coefficient of variation (53 percent) and this combined with a high sensitivity, results in the soil thickness producing over 58 percent of the variance in computed pH levels in 1980.

We conclude on the basis of the sensitivity analysis of the acidification models that there are four major parameters that determine the dynamics of long-term acidification and recovery: soil thickness, base saturation, silicate buffer rate, and the forest-filtering factor. Therefore, data on these parameters should be as reliable as possible. The initialization and parameter estimation should be based on actual field measurements; in the present applications this requirement has been only partially fulfilled.

As an example of model application, we examine acidity levels of lakes in regions of Finland, Sweden and Norway for two sulfur control scenarios for the year 2000—*30% Reduction* and *Major Sulfur Controls* (Figure 7). Output is in the form of three acidity classes of mean annual lake acidity. These classes can be set by the model user, but in this example we examine pH less than 5.3 which indicates lakes that are "strong acid dominated" (i.e. alkalinity equals zero), and pH less than 6.5 which indicates lakes that are poorly buffered but not strongly acidified.

The *Major Sulfur Controls* scenario results in significant improvement in lake acidification in southern Sweden and Finland for the lowest pH class. Differences between the two scenarios are less noticeable in other Nordic areas. This is consistent with the difference in deposition between the two scenarios (Figures 3c and 3d). This situation does not significantly change in the year 2040.

Box 6: Lake acidification submodel

Symbols:

Q_{tot}	total runoff
Q_a	quickflow (from A-layer)
Q_b	baseflow (from B-layer)
A_c	catchment area
A_l	lake area
κ_s	hydraulic conductivity
S	surface slope
Z_b	soil thickness in B-layer
Z_{tot}	total soil thickness
K_c	lumped equilibrium constant
t	time
w	catchment width
c_{HCO_3}	HCO_3^--concentration
c_H	H^+-concentration in A, B or lake (l)
wr	weathering rate
ac_f	acid load to forests
ac_o	acid load to open land
d_{tot}	total sulfur deposition
k_{SO_4}	in-lake SO_4 retention coeff.
$F_H^{(1)}$	flux of acidity from soil
$F_H^{(2)}$	flux of acidity directly on lake
$F_{HCO_3}^{(1)}$	flux of alkalinity from soil
$F_{HCO_3}^{(2)}$	flux of alkalinity from lake

Discharge from the lower soil layer (B):

$$Q_b = \kappa_s S W Z_b$$

Discharge from the upper soil layer (A):

$$Q_a = Q_{tot} - Q_b$$

Fluxes of acidity to lake:
(for calculation of concentrations see Box 5)

$$F_H^{(1)}(t) = Q_a \cdot c_{H,a}(t) + Q_b \cdot c_{H,b}(t)$$

$$F_H^{(2)}(t) = ac_o(t) \cdot A_l$$

Fluxes of alkalinity to lake:

$$F_{HCO_3}^{(1)}(t) = (wr \cdot Z_{tot} - ac_f(t))A_c$$

$$F_{HCO_3}^{(2)}(t) = \frac{k_{SO_4} d_{tot}(t)}{Q_{tot}/A_l + k_{SO_4}}$$

Equilibrium in lake mixing volume:

$$c_{H,l}(t) = \frac{K_c}{c_{HCO_3}(t)}$$

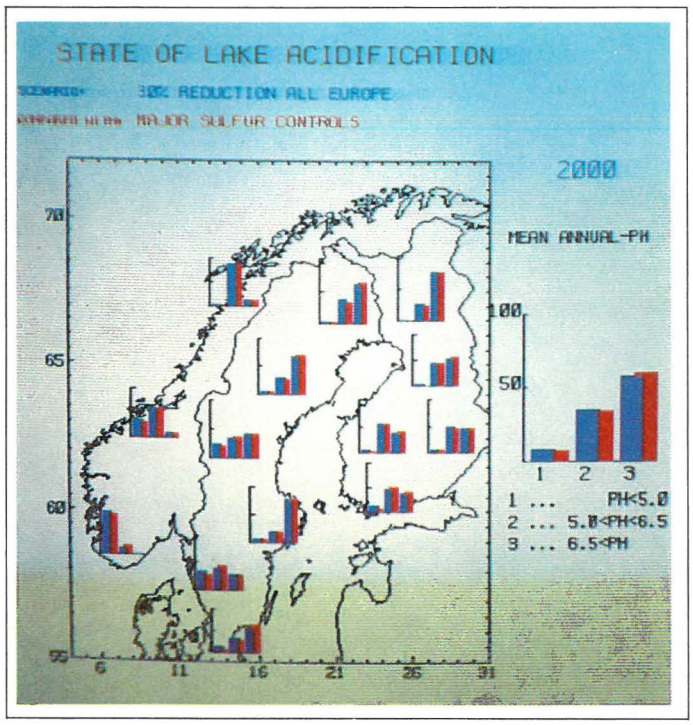


Figure 7. pH class distributions of lakes in Finland, Norway and Sweden for the *30% Reduction* (left bars) and *Major Sulfur Controls* (right bars) scenario. The bar chart at the right hand side of the picture represents the aggregated distribution for all lake regions (year 2000).

Box 7: Groundwater acidification submodel

Symbols:

bc	soil base cation content
sd	soil depth
tx	soil texture
r	recharge
as	aquifer size
am	aquifer mineral composition
f, g	qualitative functions
sens	sensitivity of groundwater
d	deposition of sulfur
risk	risk of groundwater to acidification

Sensitivity of groundwater:

$$sens = f\,(bc,\ sd,\ tx,\ r,\ as,\ am)$$

Risk of groundwater:

$$risk = g\,(sens,\ d)$$

GROUNDWATER ACIDIFICATION

The erosion of soils' natural buffering capacity by acidifying deposition, as described in the soil and lake submodels, may also lead to acidification of groundwater in Europe. Evidence of this comes from measurements of both wells and surface waters fed by goundwater (36).

The impact of acid deposition on groundwater is usually first noticed as an increasing water hardness, i.e. as calcium and magnesium are leached from the overlying soil. In areas where the soil has a low neutralizing capacity, groundwater may acidify. Where the main weathering product is aluminum, increasing levels of aluminum in groundwater may result. Increasing concentrations of sulfate coupled with a decrease in alkalinity is believed to cause corrosion of water supply pipes possibly leading to contamination of drinking water by lead and cadmium (37).

Although the hydrological and geochemical mechanisms behind groundwater acidification are qualitatively well known it is difficult to quantify the dynamic interaction between the relevant processes and the three-dimensional flow patterns on an interregional scale. We have chosen a different approach to this question (Box 7). In the initial phase, we have implemented a groundwater *sensitivity mapping system* which produces European maps of aquifer susceptibility to acidification (38).

Various factors important to groundwater acidification are compiled on a European grid: soil type, depth, and texture; aquifer size; mineral composition; and water available for recharge. The sensitivity and risk of groundwater acidification are evaluated by assessing to which extent physical and chemical soil and aquifer properties of a certain region will contribute to the neutralization of acid deposition.

Figure 8 represents typical output of the system. Northern and mountainous regions with thin soils and low weathering capability are more sensitive to groundwater acidification, whereas deep-soiled agricultural areas show the least sensitivity.

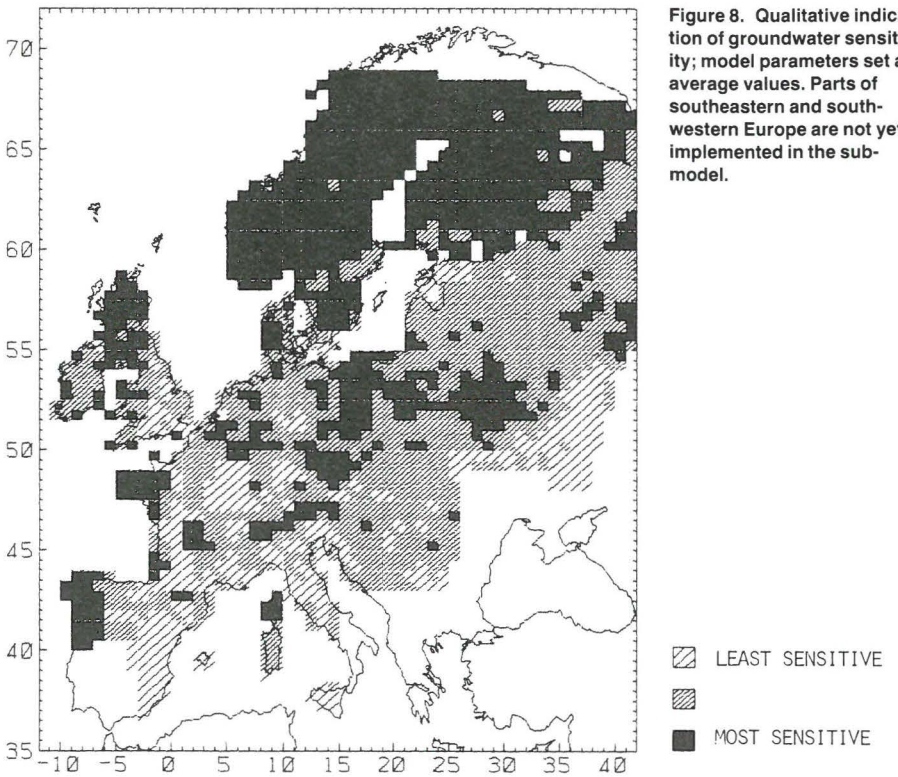


Figure 8. Qualitative indication of groundwater sensitivity; model parameters set at average values. Parts of southeastern and southwestern Europe are not yet implemented in the submodel.

DIRECT FOREST IMPACT

Forest dieback has been observed in Central Europe since the 1970s, spreading from silver fir to Norway spruce, Scots pine, and other species. Though its existence is unquestioned, its cause is in dispute. Depending on many local conditions, the following environmental stresses may be important: (1) *soil acidification* which, as noted above, can have a deleteri-

ous effect on the tree's assimilation of nutrients, (2) *direct foliar damage* due to acid deposition in which acidity erodes the protective layer of leaves; (3) *direct damage owing to elevated air concentration* of SO_2, *ozone and other pollutants* which enter the leaf tissue and affect leaf metabolism; (4) *nitrogen overfertilization*, (an excess of nitrogen deposition to the tree environment) which for example, tends to reduce frost hardiness. In reality different agents predominate in different areas, and two or more of the agents could act in concert. Also *climatic factors* and *natural stresses* almost certainly play a role in all circumstances (39).

Since we have dealt with soil acidification previously, we now consider the direct effects of SO_2. As noted, the transboundary transport of SO_2 is well established. Also the circumstantial evidence for SO_2 related forest dieback in parts of the German Democratic Republic, Czechoslovakia and Poland is rather convincing (40, 41). To quantify this forest dieback we can take three approaches: (1) *statistical/empirical models*, (2) *simulation models* of the forest environment, and (3) *indicator analysis*.

Statistical/Empirical Model

Based on empirical data of forest dieback from Czeckoslovakia's Erzgebirge (40) we have formulated a statistical/empirical model of "effective SO_2 dose" (42). The principal input to this model is the annual average air concentration of SO_2, which is taken from RAINS atmospheric transport model. The principal output is the accumulated dose of SO_2 to trees which is simple computation of concentration times exposure time (Box 8). Dose accumulates if a threshold SO_2 concentration is exceeded, and damage to trees is assumed to occur if the accumulated dose exceeds a threshold level. We account for the regional differences in tolerance of trees due to climatic conditions by making the threshold dose level a function of a variable called *effective temperature sum* (ETS). ETS is an integrated annual measure of the length and warmth of the growing season and it hence reflects the growth potential of a climatic region. It is calculated as the difference between actual temperature and threshold daily temperatures. For this calculation, each grid element is subdivided into altitude classes at 300-meter intervals. ETS is then calculated in each class using a three-dimensional interpolation routine of monthly average temperatures, together with a program to estimate the annual average ETS from that information (43). Thirty years average temperature data from 1088 weather stations in Europe are included in the interpolation routine. Forest area is similarly distributed into altitude classes in each grid element.

In Figure 9 we present preliminary calculations from this submodel for the *30% Reduction* scenario. These calculations use a threshold SO_2 concentration estimated for Norway Spruce (*Picea abies*). The map depicts the areas where the indicated percentage of forest is under risk. This map does not yet include data from the USSR and parts of Southern Europe. The areas where the risks of direct impacts of SO_2 are greatest are those located in the relatively high elevations with high SO_2 concentrations. This is because the effective temperature sums used for estimating the tolerance of the forest decrease with increasing elevation. The map seems to be in agreement with already observed damage; however it should be emphasized that it only displays damage caused by direct impacts of SO_2. For example, the Black Forest (Schwarzwald) where tree damage has been related to NO_x and oxidants does not show up in the sulfur risk areas.

Indicator Approach

In the statistical/empirical modeling approach we parameterize climatic effects and forest dynamics with surrogate variable ETS and accumulated dose. A more mechanistic model can be derived by treating forest dynamics and actual influential

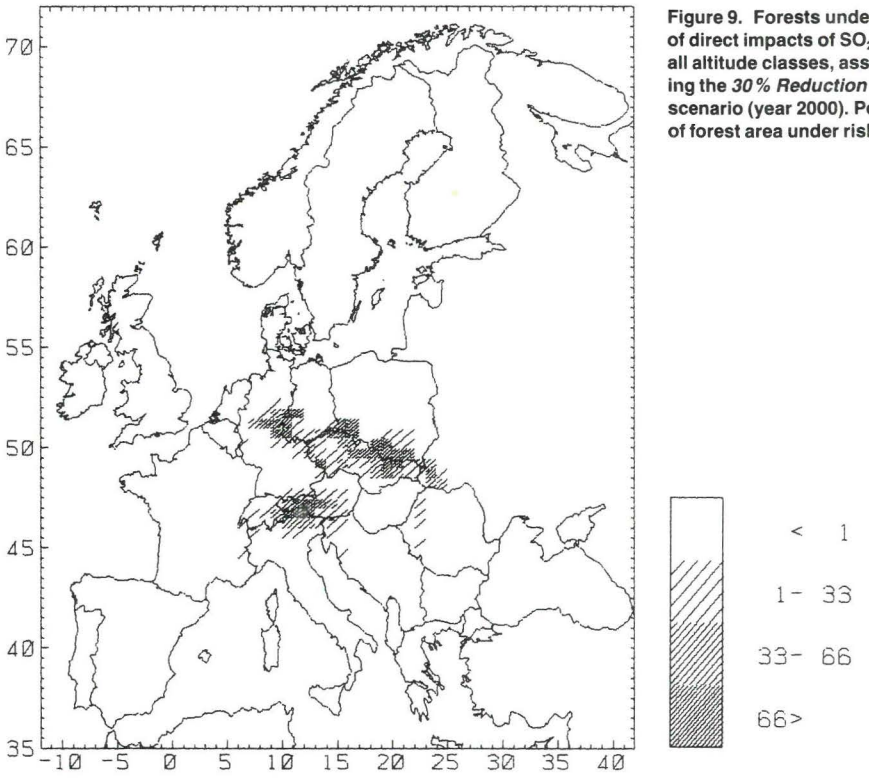


Figure 9. Forests under risk of direct impacts of SO_2 for all altitude classes, assuming the *30% Reduction* scenario (year 2000). Percent of forest area under risk.

Box 8: Forest impact submodel

Symbols:

A	annual average SO_2 air concentration
A_c	threshold SO_2 air concentration
ETS	effective temperature sum
Q	accumulated dose
Q_c	threshold accumulated dose

$$Q(t+dt) = Q(t)+f(A))dt$$

where:

$$f(A(t)) = \begin{cases} A(t) - A_c & \text{when } A(t) \geq A_c \\ 0 & \text{when } A(t) < A_c \end{cases}$$

Damage occurs when $Q(t) > Q_c$ where:

$$Q_c = f(ETS)$$

For risk assessment:

1. Determine critical time t_c from reference time t_o such that

$$Q_c = \int_{t_o - t_c}^{t_c} f(A(t))dt$$

2. *If* $t_c >$ forest rotation, then "forest under risk".

climatic variables more explicitly. This is done in the *indicator analysis approach* which is currently under development at IIASA. In this approach we use indicators of the sensitivity of forests to specified pollutant impacts, based on ecophysiological mechanisms. One group of the indicators involves synergistic impacts of air pollutants with natural stress factors, such as frost and drought (44). An indicator of increased sensitivity to drought is derived from computing the erosion of needle surfaces which is taken as a function of SO_2 concentration, temperature and fog. Similarly, the foliar concentration of sulfur is considered as an indicator of sensitivity to frost damage. The occurrence of frost and drought events is predicted with physiological models of *winter hardening* (increased resistance to low temperatures) and soil-forest hydrology. The consequent increase in probability of damage under pollutant impacts is calculated in various climatically different parts of Europe. Another type of indicator is related to how well the trees can resist the direct foliar impacts caused by pollutants, either alone or together with the natural stress factors. Combined risk of forest dieback is computed as a function of foliar damage as well as measures of tree resistance to stress.

LINKAGES BETWEEN SUBMODELS

The linkages between submodels make the RAINS model more than a loose collection of different models. Since models from one discipline are rarely designed to link with other disciplines' models it is critical to give special attention to these linkages.

Sulfur Emissions—Atmospheric Sulfur

An inherent assumption in using a fixed transfer matrix to describe sulfur transport is that the total emissions in a country may change, but their spatial distribution within a country remains the same. Of course this assumption is critical to the connection between future emissions and transport. This assumption was examined by using a probabilistic method (45). It was assumed that total country emissions were known but that each grid emission had an error of ±50 percent. For different source-receptor cases the effect of this emission error was approximately 10 to 15 percent. These model experiments indicate "compensation" by the atmosphere, i.e. the effect on deposition of overestimating emissions from one grid element is compensated by underestimating the emissions from another, so that their error averages out over a long (one year) time period. Consequently, no correction has been made in RAINS to account for this assumption of a constant spatial distribution of emissions within a country.

Atmospheric Sulfur—Soil and Lake Acidification

Sulfur deposition cannot be directly converted to acid load in soil and lake watersheds. One reason is that forest areas act as much more efficient collectors of dry sulfur deposition than open land areas because trees provide increased "collection surface" (46). We account for this so-called *filtering effect* by applying a simple *filtering factor* to computed grid-average sulfur deposition which allocates more total deposition to the forested parts of grid elements than to its open areas. Nevertheless, the total amount of deposition to the grid element computed by the atmospheric submodel is conserved. Based on a literature review, we estimated the range of the *filtering factor* as 1.1 to 3.9 (49a). Because of the lack of site-specific data, we assign a constant factor of 2 to each grid element in Europe. In Northern Europe, where wet deposition predominates, a factor of 2 may exaggerate the filtering effect, whereas in Central Europe where dry deposition has the major role in total sulfur deposition, the filtering effect may be underestimated. In a sensitivity analysis, we found that this factor is, not surprisingly, of critical importance in grids with very small forested areas (31). But considering the whole of Europe its importance is not so dramatic since, on the average, forest coverage in grids is greater than five percent.

Another reason why sulfur deposition cannot be directly converted to acid stress is that alkaline dust in the atmosphere partly compensates for the acidifying effect of sulfur deposition. Alkaline deposition strongly depends on location within Europe. One estimate is that the percentage of sulfuric acid equivalents counteracted by base cations varies from 23 percent to 44 percent in different parts of Europe (49a). A constant factor of one-third is currently used in the RAINS model to account for this alkaline deposition.

Atmospheric Sulfur—Direct Forest Impact

The EMEP model upon which the RAINS sulfur transport model is based, assumes that all sulfur is homogeneously mixed in a single vertical layer one kilometer high. While this may be a suitable assumption for the purpose of computing sulfur transport over long time and space scales, it may create bias in the computation of SO_2 related forest dieback. In reality SO_2 is sometimes homogeneously mixed in the atmospheric boundary layer, particularly when convective turbulence occurs. Otherwise, however, a vertical gradient occurs. Tests conducted with the *Direct Forest Impact* submodel have demonstrated the sensitivity of its calculations to a vertical gradient of SO_2.

UNCERTAINTY ANALYSIS

The large time and space scales treated by RAINS submodels make it difficult to test them rigorously against field data. In addition: 1. observations are often unreliable because of incorrect or inconsistent measurement techniques, 2. certain important cause-effect relations may not be readily observable as in the case of the influence of a single country's pollutant emissions on pollutant deposition at a distant receptor, 3. agreement of model output with field data does not settle the question of uncertainty when the model is used for forecasting, 4. parameters in some models may be easily "tuned" such that output closely agrees with field data, 5. it is usually impossible to assemble field data for a comprehensive range of environmental conditions.

Consequently, RAINS should be subjected to a thorough sensitivity and uncertainty analysis which complements rather than replaces model validation. As described earlier, sensitivity analysis has been carried out for the soil and lake submodels. For the SO_2 emissions and atmospheric submodels an additional "uncertainty analysis" has been applied which involves: 1. *problem formulation*, in which time and space scales of the uncertainty problem are established, 2. *inventory of uncertainties*, to collect possible sources of uncertainty in a systematic fashion, 3. *screening and ranking of uncertainties*, to set priorities for quantitative evaluations, 4. *quantitative evaluation of uncertainties* which draws on a variety of analytical techniques, and finally 5. *application to routine calculations* in which uncertainty information is used as a supplement to routine calculations.

The goal of the uncertainty analysis of the sulfur emission submodel is to estimate the uncertainty of country-scale sulfur emission calculations. From the calibration procedure it was clear that the principal source of uncertainty is sulfur content of fuels. To analyze this uncertainty we are using a modified Monte-Carlo simulation approach (47). As an example, Figure 10 depicts the computed uncertainty of emissions from the Federal Republic of Germany due to uncertain sulfur content of power plant fuel. Computation of uncertainty depends heavily, of course, on the prescribed input uncertainties. In Figure 10, for example, triangular and uniform input distributions have been used.

For the sulfur transport submodel the goal of the uncertainty analysis is to determine the uncertainty of elements of the transfer matrix by examining the original EMEP model from which the matrix is derived (48). We are interested, therefore, in determining the uncertainty of grid-based sulfur deposition related to country sulfur emissions. An uncertainty taxonomy is used to assist in the inventory of uncertainties and many of these uncertainties are eliminated *a priori* in the screening step. Different methods are used to quantify uncertainties. For example, to examine model structure uncertainty we have compared calculations of different model equations under identical meteorological conditions (24). To investigate uncertainty due to interannual meteorologic variability we have conducted matrix analyses (49). To investigate the possible impact of climate change on sulfur deposition we have conducted time series analyses (49c). For parameter uncertainty we have used, as in the sulfur emission uncertainty analysis, Monte-Carlo simulation (48).

In Table 3 we compare uncertainties from different sources. For this single source-receptor combination, these uncertainties produced a 20–30 percent variability in computed total sulfur deposition. As an example of how this information is applied in RAINS, we depict in Figure 11 the uncertainty caused by a ±25 percent error in computing total sulfur deposition. Note that the effect of a constant uncertainty

has a very strong spatial variability.

Though the uncertainty analysis is still underway, we have reached some tentative conclusions:

1. In general it is feasible not only to model the long-range transport of air pollutants but also to quantify uncertainty of model calculations.
2. In many cases model errors seem to compensate. For example, Table 3 notes that in one example the uncertainty due to interannual meteorologic variability is ±32 percent for a single country's contribution to a single receptor location. However, when all countries are included, the typical uncertainty is about ±13 percent (49b). We may conclude that fairly simple models can therefore produce good results over large time and space scales.
3. To accurately estimate the effect of parameter uncertainty on model output it is more important to know the range of the parameter uncertainty than the type of their probability distribution. Of these parameter uncertainties, mixing height and wet deposition uncertainties seem to have the greatest effect on model computations. As noted previously, the uncertainties of wet deposition parameters do not, however, seem to appreciably affect the linear relationship between sulfur emissions and deposition.

Table 3. Comparison of uncertainties in the EMEP model.

Type of Uncertainty	Uncertainty of Computed Sulfur Deposition*	Notes
Non-linearity	+27%	Bias error based on model experiments with non-linear wet deposition coefficient
Geographic distribution of emissions	±18%	90% confidence interval due to ±50% range of grid emissions
Interannual meteorologic variability	±32%	Mean relative deviation for 4 meteorologic years
Parameter estimation	±25%	90% confidence interval due ±30% parameter range

* At Illmitz Austria due to emissions from German Democratic Republic; 1980 meteorological conditions.

MODEL USE

To this point we have reviewed the basic objectives of the RAINS model, the key ideas behind each of its components, and how model uncertainties are indentified. We now examine how the model is used to evaluate European-scale control strategies.

There are two basic ways of using the model: 1. scenario analysis and 2. optimization analysis. To conduct scenario analysis the user essentially moves from top to bottom through the model as depicted in Figure 1, and first specifies an energy pathway and a control strategy. The implications of these inputs can now be studied. The user has the option of examining output from any of the submodels, e.g. sulfur emissions in a particular country or group of countries, costs of control on a country basis, sulfur deposition or SO_2 concentration at different locations in Europe or mapped for all Europe, or maps of soil acidification, lake acidification, or SO_2-related forest risk. In effect, Figures 2 through 9 make up one example of a scenario analysis. Since this is an iterative process, the user normally examines this output, and based on subjective evaluation selects an alternative energy pathway and control strategy for comparison.

In optimization analysis, the user in a sense inverts the scenario analysis procedure by starting with goals of environmental protection and having the model work "backwards" to determine a cost-effective scenario for reducing sulfur emissions in Europe to accomplish these goals. Details of the optimization analysis have been published earlier (50).

Sulfur deposition goals may be set by specifying either the maximum deposition or concentration limits for any receptor-grid element (e.g. 5 g · m^{-2} · yr^{-1} or less) (51). Reduction targets may be defined as

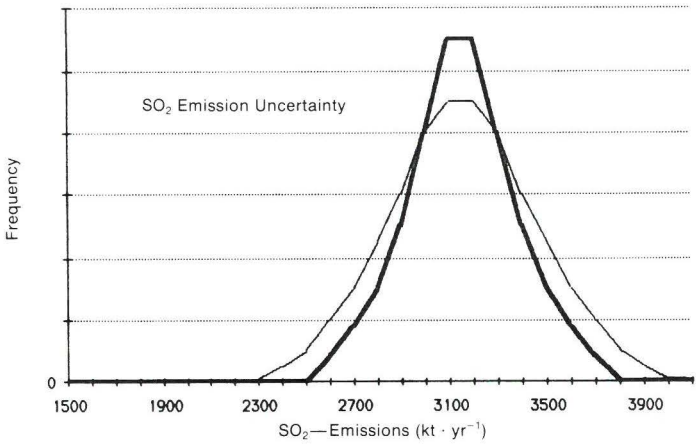


Figure 10. Computed frequency distributions of SO_2 emissions in the Federal Republic of Germany, resulting from uncertain sulfur contents of fuels used in power plants. Heavy line represents a triangular input distribution, light line a uniform distribution.

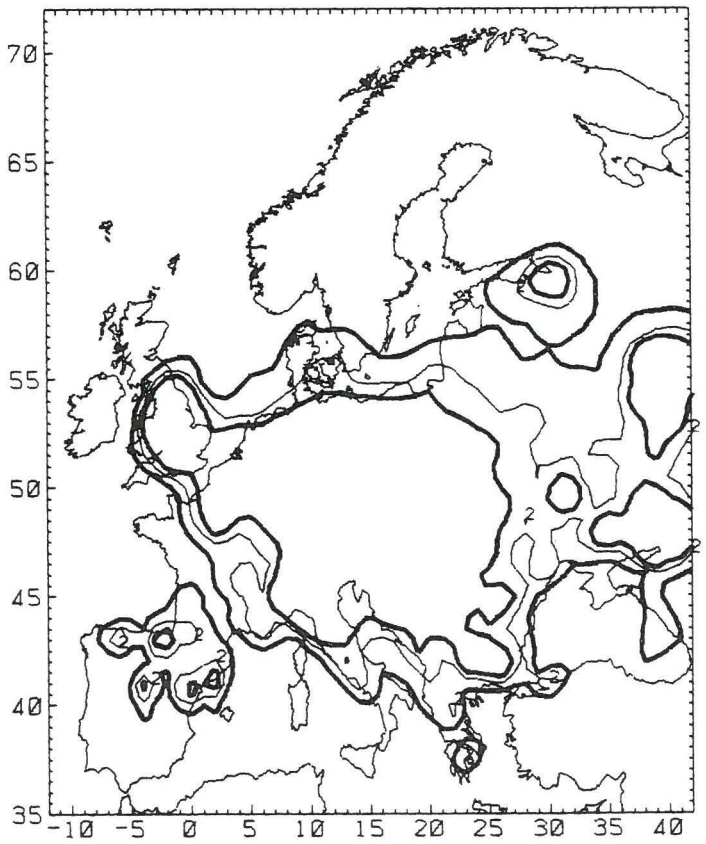


Figure 11. Computed 2 g · m^2 · yr^{-1} isoline of total sulfur deposition (light line) with ±25% uncertainty (heavy lines) assuming a *30% Reduction* scenario (year 2000).

a fraction of sulfur to be removed for each country.

Mathematically these goals are expressed in the optimization framework as constraints. (The model user should also set a series of additional constraints, such as actual limits to control technology applications, preferences on timing of control strategies, budget constraints, etc.). Once these constraints are set, the user then specifies the objective function of the optimization which can either 1. minimize total European costs, or 2. minimize total European SO_2- emission reductions. Costs calculations are based on the national cost curves described previously.

Because of computational difficulties it is undesirable to conduct the optimization analysis for every grid element. Therefore, only certain selected receptors are actually taken into account. We select these receptors so that, if a goal is met in these receptors we can be certain that the goal is met in the entire grid.

To illustrate an application of the optimization analysis, suppose we wish to limit sulfur deposition to a maximum of 5 g $\cdot$ m^{-2} $\cdot$ yr^{-1} everywhere in Europe and determine the minimum of European expenditures necessary to accomplish this, and at the same time require each country to reduce its emissions no less than (the already agreed to) 30 percent relative to their 1980 emissions. The resulting SO_2 emissions allowed for each country are given in the third column of Table 2. Note that the total emissions of this scenario in the year 2000 (12.4 MT $\cdot$ yr^{-1}) are about the same as the *Major Sulfur Controls* scenario. However, since deposition limits are obtained in a cost-effective way, our preliminary calculations indicate that total European costs of controls are significantly lower than those of *Major Sulfur Controls*. Of course, the two scenarios are not strictly comparable because their objectives are different.

Figure 3e presents the deposition from the *Deposition Limit* scenario. It is somewhat surprising that this deposition pattern is similar to that resulting from the *Major Sulfur Controls* scenario (Figure 3d) since their country-by-country SO_2 emissions differ significantly (Table 2). This "smoothing" of deposition patterns may actually occur in nature or simply be an artifact of the atmospheric submodel used in RAINS.

Since the *Deposition Limit* and *Major Sulfur Controls* scenarios produce similar deposition levels, we expect their computed environmental effects also to be similar. This is illustrated by results from the forest soil submodel in Figure 6.

SOME FINDINGS FROM USING *RAINS*

Although the RAINS model is still being developed we can summarize some preliminary conclusions from the model runs presented in this paper:

1. A concerted SO_2 emission reduction program (as in the *Major Sulfur Controls* presented in this paper) can reduce total SO_2 emissions in Europe to their 1940s level.
2. We have estimated a cost optimal reduction required for each European country to achieve a maximum of SO_2 deposition of 5 g $\cdot$ m^{-2} $\cdot$ yr^{-1} in Europe. This sums up to a 58 percent reduction of European SO_2 emissions relative to their 1980 level. In addition, this SO_2 reduction program is significantly cheaper than the *Major Sulfur Controls* scenario which accomplishes the same total European SO_2 emission reductions. Despite the different country-by-country distribution of these emissions a similar deposition pattern results from both scenarios, and consequently the two scenarios result in similar environmental effects.
3. Current emission levels result in total sulfur deposition greater than 4 g $\cdot$ m^{-2} $\cdot$ yr^{-1} throughout most of Central Europe, and parts of UK and USSR.
4. The *30% Reduction* scenario greatly reduces the area covered by deposition in the higher deposition range.
5. The *Major Sulfur Controls* scenario significantly increases the area of Europe in the lower range of deposition (less than 1 g $\cdot$ m^{-2} $\cdot$ yr^{-1}) compared to the *30% Reduction* scenario.
6. The importance of model uncertainty on deposition calculations depends very much on the level of deposition and on location. Also, model errors are found to compensate to a degree.
7. The *Major Sulfur Controls* scenario results in large improvement of soil acidification in Central Europe compared to the *30% Reduction* scenario, but small differences are observed elsewhere. There is a much greater difference between the two scenarios in the year 2040 than in 2000.
8. The *Major Sulfur Controls* scenario results in improved lake-acidification conditions in Southern Finland and Sweden compared to the *30% Reduction* scenario.
9. Because of the long-term dynamics of soil and lakes, the differences between control strategies may not be too apparent in the short term (to the year 2000), but may be more obvious in the long term. Since the ultimate goal of emission reductions in Europe is protection of the environment, the evaluation of control strategies should account for long-term effects.

OTHER USES OF *RAINS*

Apart from using it to analyze control strategies the RAINS model may be used in other ways as well:

Research priorities. From developing and testing the submodels of RAINS we have also gained insight into which data would be most useful to improve our

References and Notes

1. Whelan, T. 1986. Environmental diplomacy: the management and resolution of transfrontier environmental problems. *Ambio 15*, 56.
2. Alcamo, J.M., Hordijk, L., Kämäri, J., Kauppi, P., Posch, M. and Runca, E. 1985. Integrated analysis of acidification in Europe. *J. Environ. Manage. 21*, 47-61.
3. Hettelingh, J.P. and Hordijk, L. 1986. Environmental conflicts: the case of acid rain in Europe. *Ann. Reg. Sci. 20*, 38–52.
4. Watson, S.R. 1986. Modelling acid deposition for policy analysis. *J. Oper. Res. Soc. 37*, 893–900.
5. Chadwick, M.J. 1986. Co-ordinating economic and ecological goals. In *The Assessment of Environmental Problems*, Conway, G.R. (ed.). Imperial College, London, p. 41–49.
6. Marnicio, R.J., Rubin, E.S., Small, M.J. and Henrion, M. 1986. The acid deposition assessment model: an integrated framework for benefit and cost analysis. In *Proc. 7th World Clean Air Congress*, Sydney, Australia.
7. Balson, W.E., Boyd, D.W. and North, D.W. 1982. *Acid Deposition: Decision Framework*. Report EA-2540, Electric Power Research Institute, Palo Alto, California.
8. Meeting of Acidification Research Coordinators. 1986. Bilthoven, The Netherlands, personal communication.
9. Semb, A. 1978. Sulphur emissions in Europe. *Atmos. Environ. 12*, 455–460.
10. Ivanov, M.V. 1984. *The Global Biogeochemical Sulphur Cycle*. John Wiley, Chichester, UK.
11. UN-Economic Commission for Europe. *Energy Data Bank*. October 1986. Geneva.
12. Messner, S., Golovin, A. and Strubegger, M. 1986. *Natural Gas in Europe*. IIASA, Working Paper WP-86-39. Laxenburg, Austria.
13. Messner, S. and Strubegger, M. 1986. *First Order Effects of a Nuclear Moratorium in Central Europe*. IIASA, Working Paper WP-86-80. Laxenburg, Austria.
14. International Energy Agency. 1986. *Coal Information Report*. Paris.
15. Economic Commission for Europe. 1983. *An Efficient Energy Future*. Butterworths, London.
16. Dovland, H. and Saltbones, J. 1986. *Emissions of Sulphur Dioxide in Europe in 1980 and 1983*. EMEP-CCC Report 86/1. Lillestrom, Norway.
17. Lübkert,B. 1987. *A Model for Estimating Nitrogen Oxide Emissions in Europe*. IIASA, Working Paper. Laxenburg, Austria (in press).
18. Amann, M. and Kornai, G. 1987. *Cost Functions for Controlling SO_2 Emissions in Europe*. IIASA, Working Paper WP-87-65. Laxenburg, Austria.
19. Möller, D. 1984. Estimation of the global manmade sulfur emission. *Atmos. Environ. 18*, 19–27.
20. Fjeld, B. 1976. *Consumption of Fossil Fuels in Europe and Emissions of SO_2 During the Period 1900–72*. Norwegian Institute for Air Research. Teknisk Notat No. 1/76 (in Norwegian).
21. Rodhe, H. 1978. Budgets and turnover times of atmospheric sulfur compounds. *Atmos. Environ. 12*, 671–680.
22. Organisation for Economic Cooperation and Development. 1979. *The OECD Programme on Long Range Transport of Air Pollutants. Measurements and Findings. Paris*.
23. Eliassen, A. and Saltbones, J. 1983. Modelling of long-range transport of sulphur over Europe: a two-year model run and some model experiments. *Atmos. Environ. 17*, 1457-1473.
24. Alcamo, J., Bartnicki, J. and Schöpp, W. 1986. *Effect of non-linear sulfur removal coefficients on computed sulfur source-receptor relationships: some model experiments*. Paper presented at EURASAP Symposium on Interregional Air Pollutants Transport, Budapest, April 22–24.
25. Lehmhaus, J. Saltbones, J. and Eliassen, A. 1986. *A Modified Sulphur Budget for Europe for 1980*. EMEP/MSC-W Report 1/86, Norwegian Meterorological Institute, Oslo.
26. Asman, W.A. and Janssen, A.J. 1986. *A Long Range Transport Model for Ammonia and Ammonium for Europe and Some Model Experiments*. IMOU-Report R-86-6. Institute for Meteorology and Oceanography, Utrecht, The Netherlands.
27. Derwent, R.G. 1986. *The Nitrogen Budget for the UK and N.W. Europe*. AERE, Harwell, Oxfordshire, ETSU-R-37.
28. Ulrich, B. 1983. A concept of forest ecosystem destabilization and of acid deposition as driving force for destabilization. In *Effects of Accumulation of Air Pollutants in Forest Ecosystems*, Ulrich, B. and Pankrath, J. (eds.). Reidel, Dortrecht, The Netherlands, p. 1–29.
29. Van Breemen, N., Driscoll, C.T. and Mulder, J. 1984. Acid deposition and internal proton sources in acidification of soils and waters. *Nature 307*, 599–604.
30. Kauppi, P., Kämäri, J., Posch, M., Kauppi, L. and Matzner, E. 1986. Acidification of forest soils: model development and application for analyzing impacts of acidic deposition in Europe. *Ecol. Modelling 33*, 231–253.
31. Posch, M., Kauppi, L. and Kämäri, J. 1985. *Sensitivity Analysis of a Regional Scale Soil Acidification Model*. IIASA, Working Paper WP-85-45. Laxenburg, Austria.

understanding of acidification in Europe. For sulfur emissions, an improved description of sulfur content in fuels is needed. To improve our estimates of sulfur transport in the atmosphere it is important to describe the short time-scale dynamics of wet deposition processes and mixing-layer heights. For soil and lake acidification, the data that require special attention are base saturation and silicate buffering rate of soils, soil thickness, and the "forest filtering factor".

Data bases. RAINS also provides European-scale computerized data bases for historical energy consumption and official government projections, soil types and characteristics, forest area coverage, elevation, aquifer size, and a variety of climatic variables.

SOME FINAL REMARKS

After a description of RAINS and its use we now return to our opening remarks about the information gap between scientists and policy makers. One way the RAINS model helps to close this gap is by organizing and collecting critical scientific information about the acidification problem in a single accessible computer model. Another has been to make the graphical output of this model understandable to the policy analyst without specialized scientific training. The complexity and diversity of information needed to analyze control strategies for acidification in Europe also requires that RAINS be flexible. We have tried to build flexibility into RAINS by allowing the model user to interactively specify: 1. control strategies and other input, 2. one or several environmental impact indicators (deposition, soils, lakes, forests) for evaluation, 3. the form of output (maps, bar charts, time series, comparisons), and 4. criteria for optimization analysis. To further facilitate its use, RAINS is available on a personal computer.

As we pointed out at the beginning of this paper, one of our principal goals is to provide an overview of the acidification problem to assist in policy analysis. Unfortunately, many important details must be neglected in RAINS in order to provide this overview. These details concern connections of the European acidification problem with other European and global problems. For instance: 1. the relationship between sulfur and nitrogen pollutants and heavy metals and toxic organics, 2. the synergistic effects of pollutants in forests, 3. social factors involved with control strategies such as employment, trade balances, etc., 4. linkages with the global environment, as in the effect of fossil fuel combustion on not only sulfur and nitrogen emissions but also CO_2 emissions.

Not only do we simplify RAINS by neglecting some details of the acidification problem, but we also neglect some linkages and feedbacks between submodels that may create important nonlinear system behavior. For example, we do not take into account non-linearities that may arise because SO_2 pollution affects trees, which in turn might lose foliage and be less able to absorb SO_2, which will modify the SO_2 air concentration, which in turn will have a different effect on trees, and so on.

Considering these and other limitations of the RAINS model, one may conclude that the RAINS model should not be the only basis for decision-making. Obviously, other sources of information and analysis should also be used to select the best control strategy.

In summary, this paper presents a synthesis of some important aspects of acidification in Europe, in a quantitative rather than simply qualitative manner, and with an emphasis on large time and space scales. We have also tried to organize this information in a way useful to non-technical specialists so that science can play an even larger role in the important decisions being made to control acidification of Europe's environment.

32. National Swedish Environment Protection Board. 1986. *Acidic and Acidified Waters* (in Swedish).
33. Christophersen, N., Dymbe, L.H., Johannessen, M. and Seip, H.M. 1984. A model for sulphate in streamwater at Storgama, Southern Norway. *Ecol. Modelling 21*, 35–61.
34. Baker, L.A., Brezonik, P.L. and Pollman, C.D. 1987. Model of internal alkalinity generation: sulfate retention component. *Water, Air, Soil Pollut. 31*, 89–94.
35. Kämäri, J., Posch, M., Gardner, R.H. and Hettelingh, J. -P. 1986. *A Model for Analyzing Lake Water Acidification on a Large Regional Scale—Part 2: Regional Application*. IIASA, Working Paper WP-86-66. Laxenburg, Austria.
36. Von Brömssen, U. 1986. Acidification and drinking water—groundwater. In *Acidification and Its Policy Implications*, Schneider, T. (ed.). Elsevier, Amsterdam, p. 251–262.
37. Holmberg, M. 1986. *The Impact of Acid Deposition on Groundwater: A Review*. IIASA, Working Paper WP-86-31. Laxenburg, Austria.
38. Holmberg, M., Johnston, J. and Maxe, L. 1987. *Assessing aquifer sensitivity to acid deposition*. Presented at the International Conference on the Vulnerability of Soil and Groundwater to Pollutants. Noordwijk aan Zee, The Netherlands.
39. McLaughlin, S.B. 1985. Effects of air pollution on forests, a critical review. *J. Air Pollut. Control Assoc. 35*, 512–534.
40. Materna, J. 1985. Results of the research into air pollutants impact on forests in Czechoslovakia. *Proc. Sympos. Effects of Air Pollution on Forest and Water Ecosystems*, p. 127. Helsinki.
41. Molski, B., Bytnerowic, A. and Dmuchowski, W. 1983. Mapping air pollution of forests and agricultural areas in Poland by sulfur accumulation in pine (*P. Silvestris*). *Aquilo Ser. Bot. 19*, 326–331.
42. Mäkelä, A., Materna, J. and Schöpp, W. 1987. *Direct Effects of Sulphur on Forests in Europe—A Regional Model of Risk*. IIASA, Working Paper WP-87-57. Laxenburg, Austria.
43. Ojansuu, R. and Henttonen, H. 1983. Estimation of local values of monthly mean temperature, effective temperature sum and precipitation sum from the measurements made by the Finnish Meteorological Office. *Silva Fenn. 17*, 143–160. (in Finnish, summary in English)
44. Mäkelä, A. and Huttunen, S. 1987. *Cuticular Erosion and Winter Drought in Polluted Environments—A Model Analysis*. IIASA, Working Paper WP-87-48. Laxenburg, Austria.
45. Alcamo, J. 1987. Uncertainty of forecasted sulfur deposition due to uncertain spatial distribution of emissions. *Proc. Sixteenth International Technical Meeting on Air Pollution Modeling and Its Applications*, Lindau, FRG, April 6–10.
46. Mayer, R. and Ulrich, B. 1974. Conclusions on filtering action of forest from ecosystem analysis. *Oecol. Plant. 9*, 157–160.
47. Gardner, R.H. and Trabalka, J.R. 1985. *Methods of Uncertainty Analysis for a Global Carbon Dioxide Model*. US Department of Energy, Office of Energy Research, Report DOE/OR/21400-4. Washington, DC.
48. Alcamo, J. and Bartnicki, J. 1987. A framework for error analysis of a long range transport model with emphasis on parameter uncertainty. *Atmos. Environ. 21*, 2121–2131.
49a. Kämäri, J. 1986. Linkage between atmospheric inputs and soil and water acidification. In *Atmospheric Computations to Assess Acidification in Europe: Work in Progress*, Alcamo, J. and Bartnicki, J. (eds.). IIASA, Research Report, RR 86–5, Laxenburg, Austria.
49b. Alcamo, J. and Posch, M. 1986. Effect of interannual meteorologic variability on computed sulfur deposition in Europe. In *Atmospheric Computations to Assess Acidification in Europe: Work in Progress*, Alcamo, J. and Bartnicki, J. (eds.). IIASA, Research Report, RR 86–5, Laxenburg, Austria.
49c. Pitovranov, S. 1986. A method to assess the effects of possible climate change on sulfur deposition patterns in Europe. In *Atmospheric Computations to Assess Acidification in Europe: Work in Progress*, Alcamo, J. and Bartnicki, J. (eds.). IIASA, Research Report, RR 86–5, Laxenburg, Austria.
50. Batterman, S., Amann, M., Hettelingh, J.-P., Hordijk, L. and Kornai, G. 1986. *Optimal SO_2 Abatement Policies in Europe: Some Examples*. IIASA, Working Paper WP-86-42. Laxenburg, Austria.
51. In the current version of RAINS we do not specifically treat goals for any of the ecological models because of the technical complexity (but not impossibility) of formulating the optimization problem for nonlinear models on such large temporal and spatial scales as are covered by the model. We assume that ecological targets may also be expressed in terms of maximum sulfur deposition.

52. Special thanks are due to past and current participants in the Acid Rain Project: Jerzy Bartnicki, Stuart Batterman, Bob Gardner, Vicky Hsiung (who typed and re-typed various drafts of this paper), John Johnston, Barbara Lübkert, Lena Maxe, Sergej Pitovranov, Maximilian Posch, Wolfgang Schöpp and Miyoko Yamada. The World Meteorological Organization and the UN Economic Commission for Europe have given permission to use results from the EMEP programme. The Norwegian Meteorological Institute provided the atmospheric transfer matrices used in RAINS. Errors in this article are the authors' sole responsibility.

IIASA's model RAINS has been developed since 1983 by an interdisciplinary research team.

Joseph Alcamo initiated the emissions calculations, and is responsible for atmospheric modeling and uncertainty analysis, Markus Amman oversees energy scenarios and emission calculations, Jean-Paul Hettelingh coordinates economic research and all computer programming, Leen Hordijk is project leader, Lea Kauppi collaborated in lake and soil acidification modeling, and Pekka Kauppi coordinated the work on the forest soil acidification model. Their current address is: Acid Rain Project, International Institute for Applied Systems Analysis, A-2361 Laxenburg, Austria.

Maria Holmberg initiated the groundwater modeling, and Annikki Mäkelä led the research on modeling direct effects on forests. Their current address is: Department of Silviculture, University of Helsinki, Unioninkatu 40B, SF-00170 Helsinki, Finland.

Juha Kämäri is principal investigator of the lake acidification model. His current address is: National Board of Waters, Water Research Institute, P.B. 250, SF-00101, Helsinki, Finland.

Gabor Kornai's main responsibility is the cost-of-control model. His current address is: Institute for Economic and Market Research, Budapest, V., Dorottya u.6, H-1389, Hungary.