Methodologies for the assessment and mapping of critical loads and of the impact of abatement strategies on forest soils

W. de Vries

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ABSTRACT

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Methodologies are described for assessing and mapping critical acid loads and the impact of abatement strategies for forest soils. The various steps which are discussed are: determination of critical chemical values, selection of a computation model, collection of input data and procedures for mapping critical loads. Furthermore, the various sources of uncertainty are discussed. The computation models described are the steady-state and dynamic soil acidification models START, MACAL, SMARD and RESAM, that are the part of integrated acidification models. Major emphasis is given to the description of these models and the collection of input data to apply them on a national scale (the Netherlands) and on a European scale.

Keywords: critical load, soil acidification model, acid deposition, abatement strategies, emission scenarios.

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PREFACE

In november 1988, the "Executive Body" (EB) for the convention on "Long Range Transboundary Air Pollution" (LTRAP) of the 'United Nations - Economic Commission for Europe'(UN-ECE) has decided to install a Working Group on Abatement Strategies (WGAS). The major task of this Working Group is to assess cost-effective control strategies (emission reduction scenarios) for SO_2, NO_x and NH_3 based on existing and proposed critical loads for various receptors (forests, heathlands, crops, materials, surface waters). To accomplish this task, the Working Group is assisted by a Task Force on Integrated Assessment Modeling (TFIAM). However, the assessment of optimal control strategies requires a harmonized methodology for the assessment and mapping of both critical loads and the impact of abatement strategies in terms of ecosystem effects. This has become the task of a Working Group on Effects (WGE) that is assisted by a Task Force on Mapping (TFM). This Task Force in turn is assisted by a Coördination Centre on Critical Loads (CC).

This report describes methodologies that will be applied for forest ecosystems on a European and a national (Netherlands) scale. It includes the description of steady state and dynamic soil acidification models that are developed as part of Integrated Acidification Systems for Europe (RAINS) and the Netherlands (DAS). The research is carried out in the context of two projects at The Winand Staring Centre for Integrated Land, Soil and Water Research with the common title: "Assessment and mapping of critical acid loads on forest ecosystems and evaluation of abatement strategies". One project (7156) aims at application on a European scale and the other (7160) at application on a national scale. The information in this report is partly used in a Manual and in a Background document for Mapping critical loads of the UN-ECE Task Force on Mapping and in a Mapping Vademecum by the Coordination Centre on Critical loads.

This work was financially supported by the Dutch Ministry of Public Health, Housing and Environmental Affairs (VROM). The various models described are developed in coorperation with Ir. J. Kros (MACAL and RESAM), Dr. M. Posch, Dr. J. Kämäri and Ir. G.J. Reinds (START and SMART). Useful comments on this manuscript were given by Ir. J. Kros, Ir. B.E. Groenenberg and Ir. G.J. Reinds.

Ir. W. de Vries

SUMMARY

This report describes an overall approach for assessing and mapping critical acid loads and evaluating the impact of abatement strategies on forest soils that will be applied on a European and a National scale (the Netherlands). The major steps in the approach are discussed i.e.: the determination of critical chemical values, the selection of computation methods, the selection and area quantification of receptor types, the collection of input data and procedures for mapping critical loads. Furthermore, the various sources of error and uncertainty are discussed.

Critical chemical values for the Al-concentration, Al/Ca ratio and pH of the soil solution, related to forest decline, are given, based on insight from literature about the relation between the chemical status of soil (water) and the condition of indicator organisms such as tree species. Critical values are also given for groundwater, related to human health, and surface water, related to fish decline, since the approach can be used for these receptors as well.

The computation methods described are steady-state and dynamic (forest) soil acidification models that have been developed for application on a European and a national (the Netherlands) scale. The models are part of overall acidification simulation models that give a quantitative description of the linkages between emmission, deposition and environmental impacts such as soil acidification and effects on terrestial and aquatic ecosystems. Steady-state models predict chemical values for relevant ions in the soil solution e.g. Al, NH_4, pH etc. in an equilibrium (steady-state) situation. These models are particularly useful to derive critical loads for acid (S and N) in order to determine the final critical emission rate. The steady state models described only include processes that influence acid production and consumption during infinite time such as weathering and net nutrient uptake. A description is given of two one-layer models (SMB and START) excluding nutrient cycling, that will be applied for Europe and a multi-layer model (MACAL) including the impact of the nutrient cycle, that will be applied for the Netherlands. Dynamic models are particularly useful to predict the time scale before a critical chemical value is reached in order to determine an optimal emission scenario. The dynamic models described include processes that influence H transfer on a finite time scale, such as cation exchange, nitrogen mineralization/immobilization and sulphate adsorption/desorption. Here again, a discription is given of a one-layer model excluding nutrient cycling (SMART) and a multi-layer model (RESAM), including nutrient cycling, for application on Europe and the Netherlands respectively.

Receptors of interest are forest ecosystems, i.e. combinations of tree species and soil type. On a European scale a distinction is made between coniferous and deciduous forests whereas 81 soil types are distinguished on the basis of the 1 : 5 000 000 FAO-UNESCO Soil Map of Europe. On a national scale, a distinction is made between 12 tree species (Scotch Pine, Black Pine, Douglas Fir, Norway Spruce, Japanese Larch, Oak, Beech, Poplar, Willow, Birch, Ash and Black Alder) and 23 soil types based on a recent 1 : 250 000 Soil Map of the Netherlands. In order to apply the models, use is made of a grid with a resolution of 1.0° longitude by 0.5° latitude for the European

application and 10x10 km for the national application. Information on the area and distribution of each forest-soil combination in each grid is derived by an overlay of digitized forest - and soil databases.

For the collection of input data, emphasis is laid on the interpretation and extrapolation of available data, by deriving transfer functions (relationships) between model input data and basic land and climate characteristics such as forest type (tree species), soil type, elevation and precipitation, which are available in geographic information systems. For all the necessary input data, a data collection procedure is given. This includes atmospheric data such as deposition of SO_2, NO_x, NH_3 and base cations, hydrologic data such as precipitation, infiltration and water uptake, vegetation data such as forest growth and element contents in various tree compartments and soil data such as weathering rate, cation exchange capacity and base saturation. An overview of available data is included. Furthermore, various sampling strategies for the collection of new input data are discussed in a separate appendix.

In order to draw maps, standard procedures are discussed regarding mapping legends, mapping ranges and mapping the areal representativity of a receptor (forests). Mapping legends are given with five sensitivity classes for critical loads, excess in critical loads and various chemical criteria i.e. Al-concentration, Al/Ca ratio and pH. Procedures are given to calculate average values for each grid and to show ranges either by mapping percentile values or by giving frequency diagrams for clusters of grids.

Finally, the uncertainty in critical loads is discussed due to various sources of uncertainty, i.e.: critical chemical levels, calculation methods and input data. It is shown that especially the uncertainty in chemical criteria can have a significant impact on the critical load. Uncertainties due to assumptions in calculation methods, such as negligible N-fixation and denitrification and a simple hydrology can give rise to a high uncertainty in steep areas or in seepage areas. The uncertainty in data, either by spatial variability or because of lack of knowledge, can be quantified by an uncertainty analysis. Such an analysis will be performed on a European and a national scale in the near future.

1 INTRODUCTION

The major aim of this report, is (1) to describe models that will be used for mapping critical loads (and amounts by which they are exceeded) and for mapping the long-term impact of acid deposition on forest soils and (2) to describe strategies for the acquisition of data to apply these models on a national and a European scale. Due attention is given in this context to the separation of critical acid loads related to acidification and critical nitrogen loads related to eutrophication.

1.1 The critical load concept

Apart from direct visual damage of the forest canopy, the deposition of SO_2, NO_x and NH_3 affects forest vitality by indirect, soil-mediated effects on the roots. The most notable effect is the inhibition of base cation uptake (Ca, Mg and K), either by mobilization of Al (acidification) or by accumulation of NH_4 (eutrophication), causing unfavourable ratios of these compounds to base cations. Additional indirect effects of nitrogen include changes in vegetation due to a high nitrogen supply, increased susceptibility to frost and fungal diseases related to high leaf N contents and increased nitrate leaching to groundwater (De Vries, 1988; 1991).

In order to get derive the deposition level upon which these effects start to occur, the critical load concept has been introduced (Nilsson, 1986). The critical load on an ecosystem is defined as: "The maximum deposition of (acidifying) compounds that will not cause chemical changes leading to long term harmful effects on ecosystem structure and function" (after Nilsson and Grennfelt, 1988). A regional assessment of critical loads is very important to formulate optimal policies for emission reductions.

In order to get insight in the relation between critical loads for nitrogen, sulphur and total acid, it is important to separate between the effects of acidification, that are caused by the deposition of both sulphur and nitrogen, and eutrophication, that are caused by nitrogen deposition only. In this respect, one has to define both a critical acid load and a critical nitrogen load. The critical acid load can be defined as the maximum deposition level of both sulphur and nitrogen (total acidity) that will not cause harmfull effects on ecosystem biology due to long term acidification. Similarly, the critical nitrogen load can be defined as the maximum deposition level of nitrogen that will not cause harmful effects on ecosystem biology either by long-term eutrophication or by long-term acidification in combination with sulphur deposition.

1.2 General approach for mapping critical loads

Mapping critical loads is a laborious task involving many steps as outlined in Figure 1.

Select receptor type
↓
(Terrestrial ecosystems, groundwater, surface waters)
↓
Determine critical chemical values
↓
Select computation method (model)
↓
Quantify receptor distribution
↓
Collect input data
↓
Conduct critical load calculation
↓
Draw maps according to procedures

Fig. 1 Flowchart for mapping critical loads and areas where they have been exceeded

The various steps are:

1 Select a receptor, defined as an ecosystem of interest that is potentially impacted by atmospheric deposition, such as terrestrial ecosystems, ground-water and surface water. This report is primarily related towards forest ecosystems. A receptor is thus characterized as a specific combination of tree species and soil type. However, the methodologies described can also be used for groundwater and surface waters.

2 Define critical chemical levels, based on insight between the relation of the chemical status of soil (water), groundwater and surface water and the response of a biological indicator. A biological indicator is defined as an organism or population sensitive to chemical effects resulting from atmospheric deposition. In both forests and aquatic ecosystems, chemical effects can be related to flora and fauna (plant and animal populations). In forest ecosystems attention is generally focused on the growth and - vitality of tree species, whereas fish species are of major concern in aquatic ecosystems. In groundwater, harmful effects can be related to metal water pipes (corrosion) or humans (drinking water standards).
According to the definition in section 1.1, the critical load equals the deposition level causing a final state of a soil or lake, that does not exceed critical levels set for chemical parameters such as pH, aluminium and alkalinity. Consequently, the definition of critical chemical levels is a step of major importance in deriving a critical load.

3 Select a computation method (model). In this context, it is important to make a clear distinction between steady-state and dynamic models. Steady-state models are particularly useful to derive critical loads for total acid (S and N). These models, which only include processes that influence acid production and consumption during infinite time directly predict chemical values for relevant ions in the soil solution. Dynamic models are particularly useful to predict the time period before a critical chemical value is reached. These models also include processes that influence the acid production and consumption on a finite time scale. Summarizing, steady state models are useful to determine the final emission rate based on a final critical

acidification status, whereas dynamic models are necessary to determine an optimal emission scenario, based on the temporal evolution of the acidification status.
A steady-state and a dynamic approach for mapping sensitive forest areas are complementary, and should both be used to get insight in the area of forests under stress. Using the steady-state approach, the sensitive forest areas which are to be mapped are those where the present deposition exceeds the critical load. In the dynamic approach, the areas to be mapped are those where critical chemical values are exceeded at a certain point in time (e.g. 1990, 2010 and 2040). The flowchart given in Figure 1 is thus also applicable for mapping the impact of abatement strategies on the area exceeding critical chemical values.

4 Quantify the distribution and area of receptors. This can be done by using a suitable grid system. For forest ecosystems, the optimal method is an overlay of forest maps (including the various tree species) over soil maps, using available digitized information in geographic information systems (GIS).

5 Collect input data relevant for the model used, for all the considered receptors in all grids. This includes atmospheric data, hydrological data, vegetation data and soil data influencing the acid production and consumption in the various forest ecosystems.

6 Calculate chemical values of relevant parameters, either in a steady-state situation or as a function of time, for the various receptors in all grids. This step also includes the calculation of critical loads, the amount by which they are exceeded and the area in which they are exceeded for each grid.

7 Draw maps according to standard procedures. This includes rules on mapping resolution, mapping legends, averaging critical load values in a grid and visualization of the representativity of a receptor.

In this report the major steps of the approach to derive critical loads for forests in Europe and the Netherlands are discussed i.e.: the determination of critical chemical values (chapter 2), the computation methods (models) used (chapter 3), the selection of receptors and the quantification of its area and distribution (chapter 4), the collection of input data (chapter 5) and procedures for mapping critical loads (chapter 6). Furthermore, the various sources of error and uncertainty are discussed (chapter 7).

2 CRITICAL CHEMICAL LEVELS

2.1 Introduction

The discussion on critical chemical levels in this chapter is focused on forest soils. However, reference is also given to groundwater and surface water, since the methods described in this report are also applicable for these receptors. The criteria are focused on the Al-concentration (and pH) in relation to acidification and on the NO_3 concentration in relation to eutrophication.

Critical chemical levels for Al concentration, Al/Ca ratio, pH and NO_3 concentration in soils have been derived in relation to indirect effects influencing forest vitality such as decreased mycorrhizal frequency, root damage and inhibited nutrient uptake. A major difficulty in this respect is the influence of temporal and spatial variability in pH, Al concentrations and Al/Ca ratios on trees. For example, the definition of critical air concentrations of chemical compounds such as SO_2, NO_x and O_3 in relation to direct effects on the forest canopy is differentiated by the time of exposure. The critical concentration decreases when the time of exposure increases. The same principle holds for surface water. For example episodic events, characterized by high concentrations of Al during a short time, may be fatal to fish even though the yearly averaged concentration is not toxic. However, the insight in dose response relationships, regarding the effects of soil acidification on tree species is much less advanced. Consequently, the criteria that are given in this paragraph are taken as flux averaged yearly concentrations or ratios. Higher concentrations, which are due to temporal variability in deposition, litterfall, mineralization and water and nutrient uptake, are thus allowed during certain periods of the year. With respect to the aspect of spatial (vertical) variability, critical chemical levels are related to the forest topsoil where most of the roots occur.

2.2 Aluminium concentration

Forest soils

For forest soils exact limits for Al, above which a decreased growth and vitality of forests occurs, are difficult to define. Ulrich and his co-workers (e.g. Ulrich and Matzner, 1983), were the first to postulate the hypothesis that an increased aluminium concentration in the soil solution is a major cause of forest dieback by damaging the root system of tree species. However, a wide range of aluminium toxicity thresholds for various trees species has been reported in the literature varying between < 1.5 mg l^{-1} (0.17 $mol_c\ m^{-3}$) to > 30 mg l^{-1} (3.3 $mol_c\ m^{-3}$) (e.g. McCormack and Steiner, 1978; Steiner et al., 1980; Ryan et al., 1986a, b; Thornton et al., 1987; Smit et al., 1987; Joslin and Wolfe, 1988, 1989; Keltjens and Van Loenen, 1989). Results are all based on experiments with seedlings that were either grown in solution cultures or in a greenhouse. The tolerance to Al toxicity has generally been related to root and/or shootgrowth, and sometimes to the degree of mortality. In principle a wide range of tree sensitivity to Al can be expected as it varies as a function of solution pH, Al-speciation, Ca concentration, overall ionic strength, the form of inorganic nitrogen (NH_4 or NO_3), mycorrhizal interactions and soil moisture content. Overall research findings of the ALBIOS (Aluminium in the Biosphere) project carried out in eastern North America and northern Europe indicate that red spruce is the most sensitive tree species with statistically significant biomass reductions starting to occuring near 5 mg l^{-1} (0.55 mol_c m^{-3}) of total aluminium or 2.5 mg l^{-1} (0.28 mol_c m^{-3}) of labile (inorganic) aluminium. Other moderately sensitive species are Sugar maple, Douglas fir, Larch and European beech whereas Scotch pine, Oak and Birch relatively insensitive to Al (Cronan et al., 1989).

In order to derive a critical value for acid deposition on Dutch forest eco-systems, De Vries (1988) assumed a profile averaged inorganic Al concentration of 0.2 mol_c m^{-3} (approximately 2 mg l^{-1}) to be critical for the topsoil (0-30 cm), where most of the fine roots responsible for water and nutrient uptake occur, based on results of pot experiments with Douglas fir seedlings (Smit et al., 1987). This value is rather low, but one should be aware that it is meant as an annual average concentration. Higher values do occur at this average critical limit during summer months.

Groundwater and surface water

For groundwater, there are no real biological indicators. Critical chemical values can here be related to drinking water quality. The value used for Al in the Netherlands is 0.2 mg l^{-1} (0.02 mol_c m^{-3}). In surface waters, labile Al should be less than 0.003 mol_c m^{-3} to avoid effects on various fish species (Hultberg, 1988).

2.3 Aluminium to calcium ratio

As with Al concentrations, the critical range in molar Al/Ca ratio's in forests soils is wide, varying between 0.5 and 10 (Ulrich and Matzner, 1983; Roelofs et al., 1985). An average critical value of 1.0 was proposed by Ulrich and Matzner (1983) based on the results of Rost-Siebert (1983). Results of a correlative field study between soil solution chemistry and forest vitality (Roelofs et al., 1985) indicate a similar critical ratio. Laboratory experiments about the inhibition in the uptake of base cations by seedlings of various tree species at varying molar Al/Ca ratios also indicate a critical value of 1.0 (Boxman and Van Dijk, 1988).

2.4 pH

Forest soils

Assuming gibbsite equilibrium the critical pH can be related to the critical Al concentration according to:

$$pH = (\log KAl_{ox} - \log[Al_i]) / 3 \qquad (1)$$

Where KAl_{ox} is the dissolution constant of Al-hydroxides ($(mol\ l^{-1})^{-2}$) and $[Al_i]$ is the inorganic Al concentration ($mol\ l^{-1}$).

In most literature, KAl_{ox} is referred to as the gibbsite equilibrium constant. Actually, the dissolution of aluminium oxides and/or hydroxides is mainly confined to amorphous material in acid sandy soils (Mulder et al., 1989; de Vries et al., in prep.), whereas gibbsite is generally not found in these soils. However, it is called gibbsite, because field studies have indicated that the dissolution constant for this mineral yields a reasonable prediction of aluminium in the subsoil (Mulder and van Breemen, 1987; Cronan et al., 1986).

Taking a log KAl_{ox} of 8.0 and using the critical inorganic Al concentration of 0.2 $mol_c\ m^{-3}$ given above, this leads to a critical pH of 4.0. This is equal to an H-concentration of 0.10 $mol_c\ m^{-3}$. The value of log KAl_{ox} is based on soil solution data for eight Douglas stands at a depth of 60 cm (Kleijn and De Vries, 1987; Kleijn et al.,1989). However, in the topsoil of strongly acidified soils, such as podzols, the pH is generally less due to rate limited dissolution of Al hydroxides. Assuming equilibrium an average log KAl_{ox} value computed from soil solution data in the first 30 cm of forest soils below Douglas stands was approximately 7.0. This leads to a critical soil pH of 3.7, which is equal to 0.20 $mol_c\ m^{-3}$ of H.

Groundwater and surface water

For groundwater a value of 6.0 has been reported in literature (Sverdrup et al., 1990) whereas pH in surface water should preferably stay above 5.5 to avoid fish mortality (Hultberg, 1988).

2.5 Alkalinity

Forest soils

The alkalinity concentration [Alk] (in $mol_c\ m^{-3}$) can be defined as:

$$[Alk] = [HCO_3] + [RCOO] - [H] - [Al] \qquad (2)$$

where [RCOO] is the concentration of organic anions and [Al] is the total Al concentration.

In acid forest soils with a pH near the critical value, the HCO_3 concentration is negligible. This is not the case for the RCOO concentration. However, the critical Al concentration defined before only refers to inorganic Al, since this Al-species appears to be toxic to roots (Ulrich and Matzner, 1983). Consequently, the RCOO concentration can also be neglected assuming that RCOO is completely associated with Al. For forest soils, the critical alkalinity can thus be defined as:

$$[Alk](crit) = -[H](crit) - [Al_i](crit) \quad (3)$$

where $[Al_i]$ is the concentration of inorganic Al.

Using the critical concentrations of H and Al for forest soils given above leads to a critical alkalinity of -0.30 to -0.40 $mol_c\ m^{-3}$ for topsoils and subsoils respectively. The critical alkalinity in forest soils is thus allowed to be negative.
Groundwater and surface water

For ground- and surface water the critical alkalinity is positive, due to dissociation of CO_2 at pH levels about 5. The availibity of HCO_3 can be derived from the equilibrium between pH (H concentration) partical CO_2 pressure (pCO_2) and HCO_3 according to:

$$[HCO_3] = KCO_2 \cdot pCO_2 / [H] \quad (4)$$

where KCO_2 is the product of Henry's law constant and the first dissociation constant, and pCO_2 is the partial pressure of CO_2 in the soil (bar). Taking log KCO_2 at -7.8 and assuming an average pCO_2 of 5 mbar, the critical alkalinity for groundwater is 0.14 $mol_c\ m^{-3}$. For surface water it varies between 0.02 and 0.08 $mol_c\ m^{-3}$ with an average alkalinity of 0.05 $mol_c\ m^{-3}$. A similar range has been reported by Sverdrup (1988).

2.6 Aluminium depletion

Use of an alkalinity limit based on a critical Al concentration or Al/Ca ratio in forest soils may imply that the accepted rate of Al leaching is greater than the rate of Al mobilization by weathering of primary minerals. This causes a depletion of Al hydroxides (De Vries and Kros, 1989). This might induce an increase in Fe buffering, which in turn leads to a decrease in the availability of fosfor. Using the criterium of negligible Al depletion, the critical Al-mobilization is equal to Al weathering from primary minerals, which is strongly related to base cation weathering (cf section 3.2.2).

2.7 Nitrate concentration

Forest soils

Nitrate as such is not toxic to the root system of tree species. However, an increased N availability in forests affects the herb layer towards a shift in nitrophilous species (Hommel et al., 1990). Furthermore, it may lead to an increased N content in needles, thus increasing the risk for frost damage (Aronsson, 1980) and fungal diseases (Boxman and Van Dijk, 1988). Critical nitrate concentrations in forest soils are difficult to asses. Natural nitrate leaching rates in Central Europe are generally less than 40 $mol_c\ ha^{-1}\ yr^{-1}$ (Schulze et al., 1989), Using a precipitation surplus of 200 mm yr^{-1} (cf section 5.1.6) this leads to 0.02 $mol_c\ m^{-3}$. A similar value is given in Rosén (1990) on the basis of NO_3 concentrations in stream water of nearly unpolluted forested areas in Sweden.

However, it is questionable whether such a low value can be used, since vegetation changes occur at an increased N availability. For example, vegetation changes in Dutch heathlands can occur above N loads of 700 to 1000 mol_c ha^{-1} yr^{-1}. This implies an increased N availability of at least 300 mol_c ha^{-1} yr^{-1}. Using a precipitation surplus of at least 300 mm yr^{-1}, this gives a critical NO_3 concentration of 0.1 mol_c m^{-3}.

Groundwater

Critical chemical NO_3 concentrations in groundwater can be related to the EC drinking water standard of 0.8 mol_c m^{-3}. In the Netherlands, a target value of 0.4 mol_c m^{-3} is used.

2.8 Ammonium to potassium ratio

Roelofs et al. (1985) were among the first who postulated that increased NH_4 concentrations and ratios of NH_4 to K and Mg are an important cause of decreasing forest vitality in the Netherlands. They found a resonable correlation between the ratios of NH_4 to K and Mg in the topsoil and the vitality of coniferous trees. The accumulation of ammonium in the soil, induced by the deposition of NH_3 and NH_4, appears to inhibit the growth of ectomycorrhizae, which play an important role in nutrient uptake by many coniferous trees (Boxman et al., 1986). Imbalanced nutient concentrations in the soil solution can cause K and Mg deficiencies, resulting in chlorotic yellow-brown needles (Roelofs et al., 1985; Boxman and Van Dijk, 1988). Boxman and Van Dijk (1988) found a strong decrease in the uptake of Ca and Mg at an increasing molar NH_4/K ratio in a greenhouse experiment with two-year-old Corsican pines. Using these data Boxman et al. (1988) proposed a critical NH_4/K ratio of 5.

2.9 Summary

A summarizing overview of the various criteria that can be used for forest soils, groundwaters and surface waters is given in Table 1.

Table 1 Suggested average critical chemical levels for various parameters in forest soil, groundwater and surface water

Criteria	Unit	Forest soil	Ground-water	Surface water
[Al]	mol_c m^{-3}	0.2	0.02	0.003
Al/Ca	mol mol^{-1}	1	-	-
pH	-	4.0[1)]	6.0	5.5
[Alk]	$mol_c m^{-3}$	-0.30[1)]	0.14	0.05
NO_3	$mol_c m^{-3}$	0.10[2)]	0.8[3)]	-
NH_4/K	mol mol^{-1}	5	-	-

1) For forest topsoils, a pH value of 3.7 and an alkalinity of -0.40 mol_c m^{-3} is suggested.
2) Related to vegetation changes.
3) In the Netherlands a target value of 0.4 mol_c m^{-3} is also used.

From Table 1, it follows that for forests soils alkalinity is allowed to be negative, whereas for ground- and surface water alkalinity should be positive. This difference in critical alkalinity means that in a forested catchment the limits set for ground- and surface water generally overrule the limits set for the forest soils, except for situations where the base cation weathering in the unsaturated zone (groundwater) or in the catchment (surface water) is much larger than in the rooting zone (see also section 3.2.2).

3 MODELS

3.1 Introduction

In this chapter steady-state and dynamic soil models are described that have been developed to map sensitive terrestrial (forest) ecosystems in Europe and the Netherlands.

Steady-state models only include processes that influence acid production and consumption during infinite time. Two type of models are described: one-layer models excluding nutrient cycling (SMB, START) and a multi-layer model including nutrient cycling (MACAL). Processes considered in the one-layer models are deposition, weathering and net uptake of base cations and net uptake of nitrogen. Additional processes considered in MACAL are litterfall and nutrient uptake within the rootzone.

Dynamic models include the same processes considered in the steady state models. However, processes neutralizing the acid input on a finite time scale are also considered, such as cation exchange, nitrogen mineralization/immobilization and sulphate adsorption/desorption. Again, two models are described: a one-layer model excluding nutrient cycling (SMART) and a multi-layer model including nutrient cycling (RESAM).

The various models have been developed at the Winand Staring Centre for Integrated Land Soil and Water Research (WSC) in a joint coorperation with the International Institute for Applied Systems Analysis (IIASA), The Water and Environment Research Institute in Finland and The National Institute of Public Health and Environmental Protection (RIVM) in the Netherlands.

The dynamic models are part of integrated acidification simulation models that give a quantitative description of the linkages between emissions, deposition and environmental impacts such as soil acidification and effects on terrestrial and aquatic ecosystems. The integrated models under consideration are RAINS (Regional Acidification Information and Simulation model) that has been developed at the International Institute for Applied Systems Analysis (IIASA) for application on a European scale (Alcamo et al., 1987; 1990) and DAS (Dutch Acidification Simulation model) for application in the Netherlands (Olsthoorn et al., 1990).

3.2 The steady-state one-layer model SMB

3.2.1 Model derivation

A steady-state mass balance model (SMB) is widely used at present to calculate critical loads for total acid on a European scale (Sverdrup et al., 1990; Hettelingh and De Vries, 1991) on the basis of critical values of pH, Al concentration, Al/Ca ratio and alkalinity. Here, a derivation of this model is given.

In the SMB model, the soil is considered as one compartment equal to the thickness of the rootzone (about 30-50 cm for forests) and the critical values given in Table 1 relate to element concentrations leaching from the rootzone.

A steady state situation with respect to soil acidification implies a constant pool of exchangeable base cations (BC). Consequently, the following equation regarding fluxes of base cations should hold:

$$BC_{le} = BC_{dt} + BC_{we} - BC_{gu} \qquad (5)$$

where the subscript *le* refers to leaching, *dt* to total (wet and dry) deposition, *gu* to growth uptake and *we* to weathering. Growth uptake is the net uptake that is needed for forest growth. Base cation input by litterfall and base cation removal by maintenance uptake, that is needed to resupply base cations in leaves, is not considered here by assuming that both fluxes are equal (steady-state). Units are all equal to $mol_c\ ha^{-1}\ yr^{-1}$ (mol_c is identical to eq).

Charge balance of ions in the soil leachate fluxes requires that:

$$H_{le} + Al_{le} + BC_{le} + NH_{4,le} = SO_{4,le} + NO_{3,le} + Cl_{le} + HCO_{3,le} + RCOO_{le} \qquad (6)$$

The concentrations of OH and CO_3 are taken to be zero, which is a reasonable assumption even for calcareous soils. Defining the alkalinity leaching from the soil as (cf equation (2), section 2.5).

$$Alk_{le} = HCO_{3,le} + RCOO_{le} - H_{le} - Al_{le} \qquad (7)$$

and combining the equations (6) and (7) gives:

$$SO_{4,le} + NO_{3,le} = BC_{le} + NH_{4,le} - Cl_{le} - Alk_{le} \qquad (8)$$

Combining the equations (5) and (8) leads to:

$$SO_{4,le} + NO_{3,le} = BC_{dt} + BC_{we} - BC_{gu} + NH_{4,le} - Cl_{le} - Alk_{le} \qquad (9)$$

Since Cl is a tracer, it can be assumed that the total deposition of chloride (Cl_{dt}) equals the leaching of chloride (Cl_{le}). Furthermore, leaching of ammonium ($NH_{4,le}$) can be neglected in almost all forests ecosystems due to (preferential) uptake and complete nitrification within the rootzone. Using these assumptions, equation (9) can be rewritten to:

$$SO_{4,le} + NO_{3,le} = BC^{*}_{dt} + BC_{we} - BC_{gu} - Alk_{le} \qquad (10)$$

where BC^{*}_{dt} is the total deposition of base cations not balanced by Cl.

The deposition of both chloride and sodium is mainly regulated by seasalt input. In most countries chloride reasonable balances the sum of sodium and potassium. BC^{*}_{dt} can thus be seen as the amount of divalent base cations in deposition. These ions also play a major role weathering and in uptake. Consequently, in the rest of this report BC stands for divalent base cations. This rests on the implicit assumption that the leaching of Cl is equal to the sum of Na and K leaching.

The leaching of sulphate and nitrate can be linked to the deposition of these compounds by means of a mass balance. The sulphur balance reads:

$$S_{le} = S_{dt} - S_{gu} - S_{im} - S_{re} - S_{ad} - S_{pr} \tag{11}$$

where the subscript *im* refers to net immobilization, *re* to reduction, *ad* to net adsorption and *pr* to net precipitation.

An overview of sulphur cycling in forests by Johnson (1984) suggests that the net uptake (growth uptake), immobilization, and reduction of sulphur is generally insignificant. Adsorption (and in some cases precipitation with Al complexes) can temporarily lead to a strong accumulation of SO_4, especially in Fe- and Al-oxide rich subsurface horizons (Johnson et al., 1979, 1982). However, this phenomenon is only of temporary importance (several decades) and should not be included in a steady-state model. Consequently, the total sulphur input by deposition is assumed to equal the sulphur output by leaching, leading to an equivalent proton production. Since sulphur is completely oxidized in the soil profile, $SO_{4,le}$ equals S_{le} and consequently:

$$SO_{4,le} = S_{dt} \tag{12}$$

The nitrogen balance reads:

$$N_{le} = N_{dt} + N_{fi} - N_{gu} - N_{de} - N_{im} - N_{ad} \tag{13}$$

where the subscript *fi* refers to fixation, and *de* to denitrification. Nitrogen fixation is considered negligible in most forest ecosystems (Granhall and Lindberg, 1980), except for nitrogen-fixing species, such as red alder (Van Miegroet and Cole, 1984). Adsorption of nitrogen can also be neglected when N is available as NO_3. In most forest soils, nitrogen in leachate is strongly dominated by NO_3 (Hey et al., 1991) and therefore it is reasonable to assume complete nitrification in the rootzone (N_{le} equals $NO_{3,le}$). In the topsoil the concentration of NH_4 can be quite high and this may lead to adsorption. However, the preference of the adsorption complex for NH_4 is rather low in (acid) sandy soils (Kleijn et al., 1989). Furthermore the phenomenon is only of temporary importance, comparable to sulphate adsorption. Consequently, NH_4 adsorption can be neglected in a long-term perspective. Using these various assumptions, equation (13) can be simplified to:

$$NO_{3,le} = N_{dt} - N_{gu} - N_{de} - N_{im} \tag{14}$$

Combining the equations (10), (12), and (14) leads to:

$$S_{dt} + N_{dt} = BC^{*}_{dt} + BC_{we} - BC_{gu} + N_{gu} + N_{de} + N_{im} - Alk_{le} \tag{15}$$

A critical load for sulphur and nitrogen can be derived by aiming that it does not exceed the net input of base cations ($BC^*_{dt} + BC_{we} - BC_{gu}$) plus the removal of nitrogen by net uptake, denitrification and a critical long-term immobilization rate minus a critical leaching rate of alkalinity according to:

$$CL(S_{dt}+N_{dt}) = BC^*_{dt} + BC_{we} - BC_{gu} + N_{gu} + N_{de} + N_{im}(crit) - Alk_{le}(crit) \qquad (16)$$

where CL stands for critical load.

The critical N immobilization rate refers to the formation of stable organic N compounds in the soil (Schulze et al., 1989). The critical nitrogen immobilization rate can be derived from the total amount of nitrogen in the soil divided by the period of soil formation. However, the immobilization rate may be higher on the short term (De Vries, 1988). This is important for the assessment of interim target loads by dynamic models. However, in deriving a long-term critical load, this relatively short-term accumulation should be neglected.

By defining the total deposition of (potential) acidity (Ac_{dt}) as the sum of the total sulphur and nitrogen deposition ($S_{dt} + N_{dt}$) minus the total deposition of base cations not counteracted by chloride (BC^*_{dt}) a critical load of potential acidity can be calculated according to (Sverdrup, et al., 1990):

$$CL(Ac_{dt}) = -BC_{gu} + BC_{we} + N_{gu} + N_{de} + N_{im}(crit) - Alk_{le}(crit) \qquad (17)$$

where $CL(Ac_{dt})$ is the critical load of (potential) acidity.

The term "potential" is used since NH_3 is implicitly counted as a potential acid. This is based on the assumption that NH_4 leaching from the soil is negligible. This implies that all NH_4 coming in the system that is not retained by uptake or immobilization is leached as NO_3 (see before) with an inherent acidification. Further in this text the term "critical acid load" is used in this context.

In the Netherlands, the acid load is defined as the sum of total sulphur and nitrogen deposition minus the seasalt corrected bulk deposition of base cations. This implies that the seasalt corrected dry deposition of base cations has to be included in the critical load calculation according to

$$CL(Ac_{dt}) = BC^*_{dd} + BC_{we} - BC_{gu} + N_{gu} + N_{de} + N_{im}(crit) - Alk_{le}(crit) \qquad (18)$$

It should be noted that the use of various definitions of acid loads does not influence the amount by which critical loads are exceeded, since both present and critical load are defined similarly (see also Hettelingh and De Vries, 1991).

Although the steady state model described above has been developed for application on forest soils, it can also be used to derive critical loads for groundwater and surface water. The major difference is a change in system boundaries i.e. the rootzone for forest soils, the unsaturated zone for groundwater and a catchment for surface water (Sverdrup et al., 1990). This influences the weathering rate which is determined by the parent material, and the considered depth of the soil profile according to:

$$BC_{we} = D_{sp} \cdot \overline{\overline{BC}}_{we} \tag{19}$$

where BC_{we} is the base cation weathering ($mol_c\ ha^{-1}\ yr^{-1}\ m^{-1}$) and D_{sp} is the depth of the soil profile (m). The value of D_{sp} equals the average thickness of the rootzone, unsaturated zone or catchment depending upon the receptor. Apart from weathering, there is also a difference in critical alkalinity leaching for the various receptors due to different criteria for the critical alkalinity value (cf Table 1 and section 3.2.2).

Regarding critical loads for surface water an additional term should be added in the right hand side of equation (17) due to in-lake alkalinity generation by sulphate reduction (Schindler, 1986; Shaffer et al., 1988). However, quantification of this term on a regional scale may be difficult.

3.2.2 Critical alkalinity leaching

The critical alkalinity leaching is calculated according to:

$$Alk_{le}(crit) = PS \cdot [Alk](crit) \tag{20}$$

where PS is the precipitation surplus ($m^3\ ha^{-1}\ yr^{-1}$).

For acid forest soils HCO_3 and RCOO can be neglected and the critical alkalinity leaching can be calculated as (cf equation (3), section 2.5):

$$Alk_{le}(crit) = -PS \cdot ([Al_i](crit) + [H](crit)) \tag{21}$$

In this context, one could also use the term critical acidity and change the signs from minus to plus. Values for $[Al_i]$(crit) are given in Table 1 (cf. section 2.2). [H](crit) is related to [Al](crit) by use of equation (1) (cf section 2.4).

The precipitation surplus is calculated as:

$$PS = P - I - E_a - T_a \tag{22}$$

where P is precipitation, I is interception evaporation by the forest canopy, E_a is actual soil evaporation and T_a is actual transpiration (water uptake) in the rootzone. Instead of using the precipitation surplus, i.e. soil water draining from the rootzone, one can also use an average waterflux in the rootzone to asses the critical alkalinity leaching for a forest soil, by arguing that the critical values given in Table 1 (section 2.9) are related to the topsoil where most of the fine roots do occur (De Vries, 1991). A more flexible method is to calculate the alkalinity at any given depth using a steady-state multi-layer model which includes the variation in waterflux with depth. Such a model is described in section 3.4.

An alternative approach in forest soils is to use a critical Al/Ca ratio instead of a critical Al concentration (cf section 2.3). Denoting a critical equivalent Al/Ca ratio as RAlCa(crit), a critical level of Al leaching can be calculated according to:

$$Al_{le}(crit) = RAlCa(crit) \cdot BC^{*}_{le} \tag{23}$$

where

$$BC^{*}_{le} = BC^{*}_{dt} + BC_{we} - BC_{gu} \tag{24}$$

Use of equation (23) is based on the assumption that calcium is the dominating divalent base cation in soil water. A value that can be used for RAlCa(crit) is 1.5, i.e. a molar ratio of 1.0 (cf Table 1).

Combining the equations (21), (23) and (24) leads to the following expression for the critical alkalinity leaching:

$$Alk_{le}(crit) = -RAlCa(crit) \cdot (BC^{*}_{dt} + BC_{we} - BC_{gu}) - PS \cdot [H](crit) \tag{25}$$

Combination with equation (17) leads to

$$CL(Ac_{dt}) = (1+RAlCa(crit)) \cdot (BC^{*}_{dt}+BC_{we}-BC_{gu}) - BC^{*}_{dt} + N_{gu} + N_{de} + N_{im}(crit) + PS \cdot [H](crit) \tag{26}$$

Finally, there is also the possibility to calculate the critical alkalinity leaching by aiming at a negligible Al depletion from Al hydroxides (section 2.6). In this situation one only allows mobilization of Al from primary minerals which can be calculated according to (De Vries et al., 1989a):

$$Al_{le} = Al_{we} = r \cdot BC_{we} \tag{27}$$

where r is the stoichiometrie ratio of Al to BC in the congruent weathering of silicates (primary minerals). A reasonable average value of r is 2 (De Vries et al., 1989a; Sverdrup et al, 1990). The critical alkalinity leaching thus becomes:

$$Alk_{le}(crit) = -r \cdot BC_{we} - PS \cdot [H](crit) \tag{28}$$

Combining with equation (17) gives:

$$CL(Ac_{dt}) = (1+r) \cdot BC_{we} - BC_{gu} + N_{gu} + N_{de} + N_{im}(crit) + PS \cdot [H](crit) \tag{29}$$

3.2.3 The relation between critical loads for nitrogen, sulphur and total acid

Independent from acidification, an upper limit is set on the nitrogen deposition by the eutrophication aspect. Based on equation (14) (cf section 3.2.1) this critical N load can be derived as (cf Nilsson and Grennfelt, 1988; Schulze et al., 1989):

$$CL(N_{dt}) = N_{gu} + N_{de} + N_{im}(crit) + NO_{3,le}(crit) \quad (30)$$

where $CL(N_{dt})$ is the critical nitrogen deposition, N_{gu} is the permanent uptake of nitrogen in forest due to growth, $N_{im}(crit)$ is the critical long-term immobilization of nitrogen and $NO_{3,le}(crit)$ is a critical level of nitrate leaching.

As with alkalinity leaching, the critical nitrate leaching level is determined by the product of the precipitation surplus and a critical NO_3 concentration.

A simple description for the rate of denitrification is (Breeuwsma et al., 1987; De Vries, 1991):

$$N_{de} = fr_{de} (N_{dt} - N_{gu} - N_{im}) \quad (31)$$

where fr_{de} is the denitrification fraction (-).

Combination of equation (30), with N_{dt} is $CL(N_{dt})$, and (31) yields (De Vries, 1991):

$$CL(N_{dt}) = N_{gu} + N_{im}(crit) + NO_{3,le}(crit) / (1-fr_{de}) \quad (32)$$

The denitrification rate calculated with equation (31) is related to a critical N load. For high values of fr_{de}, this critical load can become higher than the present N load. This implies the calculation of a potential denitrification rate occurring at these circumstances. In systems where fr_{de}=1 (complete denitrification) the critical N load even becomes infinite since potential denitrification becomes infinite.

From the viewpoint of eutrophication, the critical level of nitrate leaching is determined by the critical nitrate concentration. However, from the viewpoint of acidification, the critical nitrate leaching level is determined by the critical alkalinity leaching and the level of sulphate leaching (cf equation (16) and (17) in section 3.2.1).

By combining the equations (16) and (30) a critical S load can be determined according to:

$$CL(S) = BC^*_{dt} + BC_{we} - BC_{gu} - Alk_{le}(crit) - NO_{3,le}(crit) \quad (33)$$

where $NO_{3,le}(crit)$ equals the critical nitrate leaching with respect to eutrophication

As with the critical acid load, the value of BC^*_{dt} can also be substracted from the sulphur deposition. In this case the critical S load becomes:

$$CL(S^*_{dt}) = BC_{we} - BC_{gu} - Alk_{le}(crit) - NO_{3,le}(crit) \quad (34)$$

where $S^*_{dt} = S_{dt} - BC^*_{dt}$ (35)

The critical acid load can again be derived by combining equations (32) and (34) according to:

$$CL(Ac_{dt}) = BC_{we} - BC_{gu} + N_{gu} + \left(\frac{fr_{de}}{1-fr_{de}}\right) \cdot NO_{3,le}(crit) + N_{im}(crit) - Alk_{le}(crit) \qquad (36)$$

Comparison of equation (17) and equation (36) shows that the denitrification flux in equation (17) is now defined by the critical N immobilization and NO_3 leaching level and a denitrification fraction.

When the critical load for nitrogen related to eutrophication is higher than the critical acid load, which occurs when $NO_{3,le}$(crit) is higher than - Alk_{le}(crit), a critical N load related to acidification should be used (see also De Vries, 1991).

Apart from nitrate leaching, the critical deposition level for nitrogen can also be related to nutrient imbalances between NH_4 and the base cations Ca, Mg and K. This occurs in soils where nitrification is low, at least in the topsoil. This is especially important in countries with a high input of ammonia such as the Netherlands and Denmark. Consequently, separate critical loads can be derived based on critical ratios of NH_4 to base cations such as Mg and K (Boxman et al., 1988; De Vries, 1988; De Vries, 1991).

3.3 The steady-state one-layer model START

In the previous paragraph, a simple method is given for the direct calculation of a critical acid load from a given critical Al concentration or Al/Ca ratio. Another possibility is to calculate these variables at a given deposition rate using a steady-state one-layer transport model. Such is a model is START. START is the steady state version of the model SMART (Simulation Model for Acidifications Regional Trends) which is described in section 3.5. It is specifically developed for the assessment of critical acid loads on a European scale and is part of the RAINS model (cf section 3.1).

3.3.1 Basic principle

The leading equation in START to predict the soil solution concentration of major ions is the charge balance equation:

$$[H] + [Al] + [BC^*] + [NH_4] = [NO_3] + [SO_4] + [HCO_3] \qquad (37)$$

where [] denotes the concentration in $mol_c\ m^{-3}$.

As with the SMB model [BC^*], which is equal to the concentration of Ca+Mg+K+Na-Cl, is assumed to be equal to Ca+Mg and RCOO is neglected.

For X = BC^*, SO_4, NH_4 and NO_3, the concentration is calculated according to:

$$[X] = (X_{in} + X_{int}) / PS \qquad (38)$$

where X_{in} is the sum of all input fluxes to the soil ($mol_c\ ha^{-1}\ yr^{-1}$), X_{int} is the sum of all interaction fluxes in the soil ($mol_c\ ha^{-1}\ yr^{-1}$) and PS is the precipitation surplus, calculated according to equation (22). The sum of the input and interaction fluxes is equal to the leaching flux.

The aluminium concentration is calculated from the gibbsite equilibrium equation according to (cf section 2.4; equation (1)):

$$[Al] = 3.10^{-6} \cdot KAl_{ox} \cdot [H]^3 \tag{39}$$

The value 3.10^{-6} is used for the conversion from $(mol\ l^{-1})^{-2}$ to $(mol_c\ m^{-3})^{-2}$.

Similarly the $[HCO_3]$ concentration is derived by an equilibrium with [H] according to (section 2.5; equation (4)):

$$[HCO_3] = KCO_2 \cdot pCO_2 / [H] \tag{40}$$

For acid forest soils, the HCO_3 concentration can be set to zero (section 2.5). Combination of the equations (37) - (40) gives one equation with one unknown, i.e. [H], which is solved in START by a Newton Raphson iteration procedure.

3.3.2 Input and interaction fluxes

For $X = BC^*$, SO_4, NH_4 and NO_3, the input to the soil is equal to the total deposition:

$$X_{in} = X_{dt} \tag{41}$$

For $X = BC^*$, the interaction flux in START is calculated as (cf equation (5)):

$$BC^*_{int} = BC_{we} - BC_{gu} \tag{42}$$

For $X = SO_4$, the interaction flux equals zero (cf equation (12)).

When nitrification is not complete, interactions for both NH_4 and NO_3 are calculated in START according to:

$$NH_{4,int} = -NH_{4,ni} - NH_{4,gu} - NH_{4,im} \tag{43}$$

$$NO_{3,int} = NH_{4,ni} - NO_{3,gu} - NO_{3,de} - NO_{3,im} \tag{44}$$

where the subscript *ni* stands for nitrification.

The sum of $NH_{4,int}$ and $NO_{3,int}$ (N_{int}) is (cf equation (14)):

$$N_{int} = -N_{gu} - N_{de} - N_{im} \tag{45}$$

Assuming that there is equal preference for the uptake of NH_4 and NO_3, uptake fluxes for both ions are calculated in START according to:

$$NH_{4,gu} = (NH_{4,dt} / N_{dt}) \cdot N_{gu} \tag{46}$$

$$NO_{3,gu} = (NO_{3,dt} / N_{dt}) \cdot N_{gu} \tag{47}$$

where $N_{dt} = NH_{4,dt} + NO_{3,dt}$.

Nitrification and denitrification are described in START as a fraction of the net NH_4 input and NO_3 input respectively, according to:

$$NH_{4,ni} = fr_{ni} \cdot (NH_{4,dt} - NH_{4,gu} - NH_{4,im}) \tag{48}$$

$$NO_{3,de} = N_{de} = fr_{de} \cdot (NO_{3,dt} + NH_{4,ni} - NO_{3,gu} - NO_{3,im}) \tag{49}$$

where fr_{ni} and fr_{de} are a nitrification and denitrification fraction (-) respectively.

The immobilization of NH_4 and NO_3 is calculated according to:

$$NH_{4,im} = N_{im} \cdot (NH_{4,dt}/N_{dt}) \tag{50}$$

$$NO_{3,im} = N_{im} \cdot (NO_{3,dt}/N_{dt}) \tag{51}$$

3.3.3 Calculation of critical loads

When nitrification is complete and denitrification is negligible, the critical load can be calculated by comparing the predicted alkalinity value with the critical alkalinity value according to:

$$CL(Ac_{dt}) = PL(Ac_{dt}) - CLE(Ac_{dt}) \tag{52}$$

with

$$CLE\ (Ac_{dt}) = -PS \cdot ([Alk] - [Alk](crit)) \tag{53}$$

where $CL(Ac_{dt})$ is the critical acid load, $PL(Ac_{dt})$ is the present acid load and $CLE(Ac_{dt})$ is the critical load excess for acidity.

With START it is also possible to calculate a critical load for the situation that nitrification in the rootzone is not complete. In this situation a critical load cannot directly be calculated in START according to equation (52) and (53) because nitrogen transformation processes are a function of the N deposition (cf equation (48) and (49)). N deposition thus affects the critical load. In this case, the system is solved by the same set of equations given in section 3.3.1 (equation (37)-(40)), while substituting the actual Al concentration by a critical value in the charge balance equation (equation (37)). This determines the H and HCO_3 concentration according to equation (39) and (40). The concentration of all other ions (BC^*, NH_4, NO_3 and SO_4) is determined by equation (38) in combination with the various equations describing the input and interaction fluxes (section 3.3.2). The only unknown values in this combined set of equations are the deposition of NH_x, NO_x and SO_x at critical load. In order to get these three values it is assumed that the ratios of NH_x and NO_x to total N and the ratio of N to S at critical load are equal to the present ratios. This leads to one equation with one unknown, i.e. the critical N load, which is solved iteratively. The critical acid load is calculated according to:

$$CL(Ac_{dt}) = (RSN_{dep} + 1) \cdot CL(N_{dt}) - BC^*_{dt} \qquad (54)$$

where RSN_{dep} is the ratio of the present sulphur to nitrogen deposition ($CL(S_{dt}) = RSN_{dep} \cdot CL(N_{dt})$).

Using the assumption that the present ratios of NH_x and of NO_x to N and of N to S are equal to the ratios at critical load also allows an indirect assessment of the critical load with the SMB model. This can be proven as follows.

When nitrification is incomplete, equation (10) should be rewritten by including NH_4 according to:

$$SO_{4,le} + NO_{3,le} - NH_{4,le} = BC^*_{dt} + BC_{we} - BC_{gu} - Alk_{le} \qquad (55)$$

For SO_4, the leaching flux in START is calculated as (cf equation (12)):

$$SO_{4,le} = S_{dt} \qquad (56)$$

The NH_4 leaching flux, which is equal to the sum of input and interaction fluxes is calculated by combining equations (39), (43), (46), (48) and (50):

$$NH_{4,le} = (1 - fr_{ni}) \cdot \left(\frac{NH_{4,dt}}{N_{dt}}\right) \cdot (N_{dt} - N_{im} - N_{gu}) \qquad (57)$$

The NO_3 leaching flux is calculated by combining equation (39), (44), (47), (48), (49) and (51):

$$NO_{3,le} = \left(\frac{NO_{3,dt} + fr_{ni} \cdot NH_{4,dt}}{N_{dt}}\right) \cdot (N_{dt} - N_{im} - N_{gu}) - N_{de} \qquad (58)$$

Combining the equations (55) - (58) gives:

$$S_{dt}+(N_{dt}-N_{im}-N_{gu}) \cdot \left(\frac{NO_{3,dt} + (2\, fr_{ni}-1) \cdot NH_{4,dt}}{N_{dt}}\right) = BC^*_{dt}+BC_{we}-BC_{gu}+N_{de}-Alk_{le} \qquad (59)$$

Equation (59) contains three unknowns, i.e. the deposition of SO_4, NO_3 and NH_4 at critical load. Using the assumption given before, it is possible to assess a critical load. If $fr_{ni} = 1$ (complete nitrification) equation (58) is equal to equation (15) again.

3.4 The steady-state multi-layer model MACAL

3.4.1 Introduction

Runoff and leachate concentrations that are important for surface water quality are generally unaffected by the nutrient cycle. However, this is not the case for the soil water quality within the rootzone. Especially the Al/Ca ratio strongly increases with soil depth since the Ca concentration decreases with depth due to uptake, whereas the Al concentration increases due to a decrease in waterflux with depth. In areas with a relatively low precipitation surplus, this effect may be important.

Since the critical values for forest soils given in Table 1 are related to average. concentrations or ratios within the rootzone, it is better to include the effects of water uptake and nutrient cycling (foliar uptake, foliar exudation, litterfall, mineralization and root uptake). Regarding nutrient cycling, the effect of root turnover can also be included but this process generally takes place within the rhizosphere and hardly affects the yearly average concentrations or ratios in the soil solution.

Inclusion of water uptake and nutrient cycling in a steady-state approach will always lead to higher critical loads on forests, compared to those derived by a steady-state approach excluding these processes. The reason for this is that the average Al concentration and Al/Ca ratio in the rootzone is always lower than the concentration or ratio in soil water draining from the rootzone as indicated above. Average critical loads on forests in the Netherlands will therefore also be derived by an approach including nutrient cycling using a modified version of a model called MACAL (Model to Assess a Critical Acid Load; De Vries, 1988).

3.4.2 Basic principle

MACAL is not a multi-layer model in the strict sense, since the concentrations are calculated at each depth. As with START, the MACAL model is based on the charge balance principle according to:

$$[H]+[Al]+[Ca]+[Mg]+[K]+[Na]+[NH_4] = [NO_3]+[SO_4]+[Cl]+[HCO_3] \qquad (60)$$

Unlike START, the base cations are not lumped and consequently chloride is included as well.

Analogous to START, for X = Ca, Mg, K, Na, NH_4, NO_3, SO_4 and Cl, the concentration at each depth z is calculated as (cf equation (22) and (38):

$$[X](z) = \frac{X_{in} + X_{int}(z)}{P - I - E_a - T_a(z)} \tag{61}$$

The Al concentration at each depth (z) is calculated as (cf equation (39)):

$$KAl_{ox}(z) = 3.10^{-6} \cdot [H](z)^3 \tag{62}$$

The dependence of 3.10^{-6} with depth is calculated as:

$$KAl_{ox}(z) = 10^{\alpha+\beta\cdot\log(z)} \qquad \text{for } z < 100 \text{ cm} \tag{63}$$

$$KAl_{ox}(z)\ ^{\alpha+2\beta} \qquad \text{for } z \geq 100 \text{ cm} \tag{64}$$

The HCO_3 concentration at each depth is calculated as (cf equation (40)):

$$[HCO_3](z) = KCO_2 \cdot p\ CO_2 / [H](z) \tag{65}$$

Combination of the equations (60) to (65) yields one unknown i.e. [H](z), which is solved iteratively.

At the depth of the rootzone, the effect of water and nutrient cycling is negligible and the MACAL model becomes completely equal to the START model described in section 3.3, provided that KAl_{ox} at the depth of the rootzone is taken equal for both models.

3.4.3 Input fluxes

Unlike START, the element input in MACAL does not only include deposition, but also the effects of nutrient uptake or exudation by the forest canopy and the input by mineralization. In MACAL, it is assumed that these processes only affect the input of Ca, Mg and K (foliar exudation and mineralization), NH_4 (foliar uptake and mineralization), NO_3 (foliar uptake) and SO_4 (mineralization and foliar uptake). Canopy interactions and (net) mineralization fluxes of Na and Cl are generally small and have been neglected. For these ions, the input is equal to the total deposition. Since MACAL is a steady state model, mineralization is put equal to litterfall, except for NH_4, where a critical N immobilization is substracted (comparable to START and SMB). In MACAL N immobilization is assumed to occur in the litter layer above the mineral soil in the form of NH_4.

The input fluxes of Ca, Mg, K, NH_4, NO_3 and SO_4 are thus calculated as:

$$X_{in} = X_{dt} + X_{fe} + X_{lf} \tag{66}$$

$$SO_{4,in} = SO_{4,dt} + S_{lf} - SO_{4,fu} \tag{67}$$

$$NH_{4,in} = NH_{4,dt} - NH_{4,fu} + N_{lf} - N_{im}(crit) \tag{68}$$

$$NO_{3,in} = NO_{3,dt} - NO_{3,fu} \tag{69}$$

where X stands for Ca, Mg or K and the subscripts *fu*, *fe* and *lf* refer to foliar uptake, foliar exudation and litterfall respectively.

Foliar uptake and foliar exudation

Foliar uptake of NH_4, NO_3 and SO_4 is described as a linear function of total deposition.

$$X_{fu} = frX_{fu} \cdot X_{dt} \tag{70}$$

where frX_{fu} is the foliar uptake fraction of element X (-).

Foliar exudation of the cations Ca, Mg and K is assumed to be triggered by exchange with NH_4 (Roelofs et al., 1985) and H (Ulrich en Matzner, 1983) according to:

$$Ca_{fe} + Mg_{fe} + K_{fe} = NH_{4,fu} + H_{fu} \tag{71}$$

As with NH_4, foliar 'uptake' of H is assumed to be a linear fraction of the total H deposition, according to:

$$H_{fu} = frH_{fu} \cdot H_{dt} \tag{72}$$

The deposition of free H is calculated from the charge balance:

$$H_{dt} = SO_{4,dt} + NO_{3,dt} + Cl_{dt} - Ca_{dt} - Mg_{dt} - K_{dt} - Na_{dt} - NH_{4,dt} \tag{73}$$

In MACAL, the uptake fraction for H and NH_4 deposition is taken equal. Both ions are assumed to have equal preference for exchange with base cations in the forest canopy. This implies that a decrease in NH_4 deposition, which is compensated by an increase in H deposition (cf equation (73)) does not influence the foliar exudation flux of base cations. This flux is mainly triggered by a decrease or increase in SO_4 or NO_3 deposition. This can also be shown by combining the equations (70) to (73), with $frH_{fu} = frNH_{4,fu}$, which yields:

$$Ca_{fe} + Mg_{fe} + K_{fe} = frH_{fu} \cdot (SO_{4,dt} + NO_{3,dt} - BC^*_{dt}) \tag{74}$$

where BC^*_{dt} equals the deposition of the sum of base cations minus chloride (see also section 3.2.1). The deposition flux of BC^* is less liable to change than the SO_4 and NO_3 deposition.

The foliar exudation of each individual cation is calculated as:

$$X_{fe} = frX_{fe} \cdot (Ca_{fe} + Mg_{fe} + K_{fe}) \tag{75}$$

where frX_{fe} is the foliar exudation fraction of Ca, Mg or K (-).

The sum of $frCa_{fe}$, $frMg_{fe}$ and frK_{fe} equals 1.

Litterfall

The litterfall of Ca, Mg, K and S is described as:

$$X_{lf} = k_{lf} \cdot Am_{lv} \cdot ctX_{lv} \tag{76}$$

where k_{lf} is a litterfall rate constant (yr^{-1}), Am_{lv} is the amount of leaves or needles (kg ha^{-1}) and ctX_{lv} is the content of element X in leaves ($mol_c\ kg^{-1}$).

For $N(NH_4)$, litterfall is described as:

$$N_{lf} = (1 - fr_{re}) \cdot (k_{lf} \cdot Am_{lv} \cdot ctX_{lv}) \tag{77}$$

where fr_{re} is a reallocation factor (-).

Reallocation of N from the older needles to younger needles generally takes place before litterfall. However, at high deposition levels, such as in the Netherlands, reallocation of N hardly occurs (Oterdoom et al., 1987).

Consequently, the reallocation fraction is described in MACAL as a function of the N content according to:

$$fr_{re} = fr_{re,max} \cdot \left(\frac{ctN_{lv,max} - ctN_{lv}}{ctN_{lv,max} - ctN_{lv,min}}\right) \tag{78}$$

where $ctN_{lv,max}$ is a maximum nitrogen content in leaves (%) above which reallocation is nihil and $ctN_{lv,min}$ is a minimum nitrogen content in leaves below

which reallocation is at its maximum. Reallocation of Ca, Mg, K and S is generally small and is not included in MACAL.

3.4.4 Interaction fluxes

As with START, the interaction fluxes for base cations, SO_4, NH_4 and NO_3 in MACAL are base cation weathering, root uptake, nitrification and denitrification.

For the base cations, (X = Ca, Mg, K and Na) the interaction flux is described as (cf equation (44)):

$$X_{int}(z) = X_{we}(z) - X_{ru}(z) \tag{79}$$

where the subscript *ru* stands for root uptake. For Na, root uptake is zero, since this element is not included in the nutrient cycle.

For SO_4, the interaction flux equals root uptake. For NH_4 and NO_3, the interaction fluxes are described in MACAL as (cf equation (43) and (44)):

$$NH_{4,int}(z) = -NH_{4,ni}(z) - NH_{4,ru}(z) \tag{80}$$

$$NO_{3,int}(z) = NH_{4,ni}(z) - NO_{3,ru}(z) - NO_{3,de}(z) \tag{81}$$

An overview of the description of the various soil interactions in MACAL is given below.

Base cation weathering

Base cation (X = Ca, Mg, K, Na) weathering is described in MACAL as:

$$X_{we}(z) = f_{we,rz}(z) \cdot X_{we,rz} + f_{we,ss}(z) \cdot X_{we,ss} \tag{82}$$

where $X_{we}(z)$ is the cumulative weathering flux at depth z ($mol_c\ ha^{-1}\ yr^{-1}$), $X_{we,rz}$ and $X_{we,ss}$ is the weathering flux per meter soil in the rootzone and subsoil respectively ($mol_c\ ha^{-1}\ yr^{-1}\ m^{-1}$) and $f_{we,rz}(z)$ and $f_{we,ss}(z)$ is the weathering factor in the rootzone and subsoil at depth z respectively (m).

Use of equation (82) implies a separate description for weathering in the rootzone, where most of the soil formation occurs, and the subsoil wich generally consists of the parent material. When MACAL is applied to forest ecosystems, the soil depth is restricted to the rootzone. However, MACAL can also be used to assess the critical loads for phreatic groundwater. In that case, soil depth is restricted to the unsaturated zone.

The weathering pattern in the rootzone ($z = \leq DRZ$) is assumed to be non-linear and is described as:

$$fr_{we,rz}(z) = (1 - (\frac{DRZ - z}{DRZ})^m) \cdot DRZ \tag{83}$$

where DRZ is the depth of the rootzone (m) and m is a dimensionless exponent.

When $z > DRZ$, $f_{we,rz}(z) = DRZ$

The weathering pattern in the subsoil ($z > DRZ$) is assumed to be linear and is described as:

$$f_{we,ss}(z) = z - DRZ \tag{84}$$

When $z \leq DRZ$, $f_{we,ss}(z) = 0$

Root uptake

Unlike START the root uptake of the base cations Ca, Mg and K in MACAL not only consists of net uptake for forest growth, but also of maintenance uptake to resupply these base cations to the forest canopy. Since MACAL is a steady-state model, the maintenance uptake of base cations is equal to the input by litterfall and foliar exudation and the root uptake flux is described as:

$$BC_{ru}(z) = fr_{ru}(z) \cdot (BC_{lf} + BC_{fe} + BC_{gu}) \tag{85}$$

where $BC_{ru}(z)$ is the cumulative root uptake flux at depth z ($mol_c\ ha^{-1}\ yr^{-1}$) and $fr_{ru}(z)$ is the cumulative uptake fraction at depth z (-).

As with the base cations Ca, Mg, K, root uptake of nitrogen consists of net uptake for forest growth and maintenance uptake, which is equal to the input by litterfall minus foliar uptake:

$$N_{ru} = N_{lf} - NH_{4,fu} + N_{gu} \tag{86}$$

Analogous to START, the root uptake of NH_4 and NO_3 is calculated as (cf equations (46) and (47):

$$NH_{4,ru}(z) = fr_{ru}(z) \cdot N_{ru} \cdot (NH_{4,in} / N_{in}) \tag{87}$$

$$NO_{3,ru}(z) = fr_{ru}(z) \cdot N_{ru} \cdot (NH_{4,in} / N_{in}) \tag{88}$$

where N_{in} is the sum of the NH_4 and NO_3 input, as defined in the equations (68), and (69).

The root uptake of SO_4, which equals the litterfall minus foliar uptake, is calculated as:

$$SO_{4,ru}(z) = fr_{ru}(z) \cdot (S_{lf} - SO_{4,fu}) \tag{89}$$

As with weathering, the nutrient uptake pattern in the rootzone ($z \leq DRZ$) is described as (De Vries, 1988):

$$fr_{ru}(z) = 1 - \left(\frac{DRZ - z}{DRZ}\right)^n \tag{90}$$

where n is a dimensionless exponent.

For z > DRZ, $fr_{ru}(z)$ equals 1.

For n=1 the uptake pattern is uniform, for n=2 it is linear, for n=3 it is quadratic etc. The value of n will be influenced by the root distribution with depth.

The water uptake pattern is described in a similar way according to:

$$T(z) = fr_{ru}(z) \cdot T_a \qquad (91)$$

where T(z) is the cumulative transpiration (water uptake by roots) at depth z.

Nitrogen transformations

Nitrification and denitrification are described in MACAL according to (cf equation (48) and (49)):

$$NH_{4,ni}(z) = fr_{ni}(z) \cdot (NH_{4,in} - NH_{4,ru}(z)) \qquad (92)$$

$$NO_{3,de}(z) = fr_{de}(z) \cdot (NO_{3,in} + NH_{4,ni}(z) - NO_{3,ru}(z)) \qquad (93)$$

where $NH_{4,ni}(z)$ and $NO_{3,de}(z)$ are the cumulative nitrification and denitrification flux at depth z ($mol_c\ ha^{-1}\ yr^{-1}$) and $fr_{ni}(z)$ and $fr_{de}(z)$ are the cumulative nitrification and denitrification fraction at depth z (-) respectively.

As with weathering and uptake in the rootzone, the nitrification and denitrification pattern is described according to:

$$fr_{ni}(z) = fr_{ni,in} + (1 - fr_{ni,in}) \cdot (1 - (\frac{D_{ni} - z}{D_{ni}})^p) \cdot fr_{ni,rz} \qquad (94)$$

$$fr_{de}(z) = (1 - (\frac{D_{de} - z}{D_{de}})^q) \cdot fr_{de} \qquad (95)$$

where $fr_{ni,in}$ is the nitrification fraction related to the occurence of nitrification above the mineral soil (in the litter layer), $fr_{ni,rz}$ is the nitrification fraction related to the occurence of nitrification in the rootzone, D_{ni} and D_{de} are the depths over which nitrification and denitrification does occur and p and q are dimensionless exponents. Nitrification above the mineral soil is included since nitrification is an important process in the litter layer (which is not explicitly accounted for in MACAL).

For $z > D_{ni}$ and $z > D_{de}$, $fr_{ni}(z)$ and $fr_{de}(z)$ are equal to $fr_{ni,in} +(1 - fr_{ni,in}) \cdot fr_{ni,rz}$ and fr_{de} respectively, which are the total nitrification and denitrification fractions respectively, as defined before.

3.4.5 Calculation of critical loads

The critical load at a given depth z cannot be derived directly in MACAL from a comparison of the present and critical alkalinity at that depth (cf equation (52) and (53). The reason is that processes such as foliar uptake, foliar exudation, litterfall, nitrification and denitrification are a function of the deposition of SO_4, NH_4 and/or NO_3 as given before (section 3.4.3). This affects the concentration of Al and the molar Al/Ca ratio (the alkalinity) in a non-linear way.

Analogous to START, the system is solved by the set of equations given in section 3.4.2. while substituting the actual Al concentration by a critical value in the charge balance equation (equation (60)). This determines the H and HCO_3 concen-tration (alkalinity) according to the equations (62) and (65). The concentration of all other ions is determined by equation (61) in combination with the various equations uin section 3.4.3 and 3.4.4 describing the various input and interaction fluxes. The only unknown values in the combined set of equations are the critical load for NH_3, NO_x and SO_2. As with START, it is assumed that the ratios of NH_3 and NO_x to total N and the ratio of N to S at critical load are equal to the present ratios in order to calculate a critical acid load (cf section 3.3.4).

3.5 The dynamic one-layer model SMART

3.5.1 Introduction

A dynamic approach of mapping sensitive terrestrial and aquatic ecosystems, requires a model which predicts the pH, Al concentration, Al/Ca ratio etc. as a function of time. Such a model has been developed at IIASA in a joint cooperation with the Winand Staring Centre for integrated land, soil and water research in the Netherlands and the Water and Environmental Research Institute in Finland (De Vries et al., 1989a,b). Apart from the concentrations of Al, BC, HCO_3, SO_4, NO_3 and NH_4 and the pH, the model called SMART (Simulation Model for Acidification's Regional Trends) also predicts the base saturation, as a function of time (De Vries et al., 1989a,b).

SMART is the dynamic version of START. The model is specially developed in order to get insight in the impacts of different emission scenarios on forest soils in Europe. Consequently, SMART will be applied on a European Scale within the overall framework of RAINS (Regional Acidification Information and Simulation model). Moreover, national applications of the model are planned for Finland as part of the Finnish Integration Acidification model HAKOMA (Johansson et al., 1988). Apart from forest soils, SMART can also be used to predict the effects of acid deposition on surface waters in a dynamic way. In this context the model has been coupled to a lake model developed by Kämäri (1988) and applications are underway for analyzing lake water acidification on a large regional scale (Ferro scandia).

Most of the assumptions that are made to derive a critical acid load using SMB or START are also made in SMART i.e. (cf section 3.2.1):
- uptake, immobilization, reduction and precipitation of SO_4 is negligible;
- nitrogen fixation and adsorption of NH_4 is negligible.

Other assumptions made in the model are (De Vries et al., 1989a,b):
- the soil is a homogeneous compartment of constant density;
- the element input mixes completely within the soil compartiment;
- the waterflow is stationary on a yearly basis (model time step); and
- the waterflux percolating from the soil compartment equals the precipitation minus evapotranspiration.

The major difference between SMART and START is the inclusion of cation exchange, dynamic nitrogen immobilization, and sulphate adsorption, which may play an important role during a limited timeperiod. Especially cation exchange might be very important, e.g. in loamy soils with a high CEC and a high base saturation. SMART also includes carbonate weathering. The effect of this process is only implicitly accounted for in START, by using an extremely high weathering rate for calcareous soils. By doing so, the present deposition will never exceed the critical acid load on calcareous soils. In the long run (100-1000 years) both the static and the dynamic approaches will lead to the same results in non-calcareous soils, since nitrogen immobilization, cation exchange and sulphate adsorption only play a temporary role.

3.5.2 Basic principle

SMART consists of a set of mass balance equations, which describe the soil input-output relationships for the cations (Al, BC, NH_4) and strong acid anions (SO_4, NO_3), and a set of equilibrium equations, which describe the equilibrium soil processes, determining H, Al and HCO_3. As with START the concentration of Al and HCO_3 is determined by equilibrium equations (equation (39) and (40)) and the concentration of H by the charge balance equation (equation 37). As with START the concentration of base cations not balanced by chloride (BC^*) is taken to be equal to the concentration of the divalent base cations Ca + Mg.

For each of the cations (Al, BC, NH_4) and anions (SO_4, NO_3) considered in the model the mass balance equation is given by (cf equation (38)):

$$\frac{d}{dt} X_{tot} = X_{in} + X_{int} - PS \cdot [X] \qquad (96)$$

where X_{tot} is the total amount of element X in the soil ($mol_c\ ha^{-1}$) and [X] is the equivalent concentration of element X in soil water ($mol_c\ m^{-3}$).

An overview of the processes included in SMART in relation to the various ions considered is given in Table 2.

Table 2 Overview of the ions and processes included in SMART ("+" = ion included in the respective process, "-" = ion not included)

Process	H	Al	BC	NH_4	NO_3	SO_4	HCO_3
Deposition[1)]	+	-	+	+	+	+	-
Growth uptake[1)]	+	-	+	+	+	-	-
Nitrogen immobilization[2)]	+	-	-	+	+	-	-
Nitrification[1)]	+	-	-	+	+	-	-
Denitrification[1)]	+	-	-	-	+	-	-
Dissociation/association[1)]	+	-	-	-	-	-	+
Carbonate weathering	+	-	+	-	-	-	+
Silicate weathering[1)]	+	+	+	-	-	-	-
Al hydroxide weathering[1)]	+	+	-	-	-	-	-
Cation exchange	+	+	+	-	-	-	-
Sulphate adsorption	+	-	-	-	-	+	-

1) These prosesses are also included in START
2) This process is included in START as a model input

The total amount of element X in the soil is equal to the amount in the solid phase (in organic matter, in minerals and at the adsorption complex) and in the the soil solution. The amount in the solid phases is derived from the element content in these phases (mol_c kg^{-1}) multiplied by the bulkdensity r ($kg\ m^{-3}$) and the soil thickness T (m). The model contains a mass balance for carbonates, for N in organic matter, Al in hydroxides and cations and anions at the adsorption complex (Al, BC, SO_4).

3.5.3 Input an interaction fluxes

As with START, the input fluxes for X = BC^*, NO_3, NH_4 and SO_4 are equal to the total deposition (equation (41)) and the interactions of NH_4 and NO_3 in the mineral soil are described by the equations (43) to (51) (cf section 3.3.2).

Additional process descriptions included in SMART are dynamic nitrogen immobilization, carbonate weathering, cation exchange and sulphate adsorption (cf Table 2). A description is given below.

The description of nitrogen immobilization is based on the assumption that the amount of organic matter is in an equilibrium state. Consequently, immobilization of base cations is not accounted for. N immobilization only occurs by an increase in N content in organic matter. When the C/N ratio of litter is above a critical ratio ($C/N_{lt} > C/N_{cr}$), all excess nitrogen is assumed to immobilize according to:

$$N_{im} = N_{dt} - N_{gu} \tag{97}$$

Between a critical and a maximum C/N ratio ($C/N_{cr} > C/N_{lt} > C/N_{m}$), the immobilization rate is assumed to decrease according to:

$$N_{im} = (N_{dt} - N_{gu}) \cdot \left(\frac{C/N_{cr} - C/N_{lt}}{C/N_{cr} - C/N_{m}}\right) \tag{98}$$

Below C/N_m, N_{im} equals zero. The distribution of N immobilization over NH_4 and NO_3 is calculated according to equation (50) and (51).

The dissolution of calcium carbonate (e.g. calcite) is calculated according to:

$$[Ca] \cdot [HCO_3]^2 = KCa_{cb} \cdot pCO_2 \tag{99}$$

where KCa_{cb} is the equilibrium constant for calcium carbonate dissolution.

The various exchange reactions are described by Gaines-Thomas equations using concentrations instead of activities:

$$\frac{frH^2{}_{ac}}{frBC_{ac}} = KH_{ex} \cdot \frac{[H]^2}{[BC]} \tag{100}$$

$$\frac{frAl^2{}_{ac}}{frBC^3{}_{ac}} = KAl_{ex} \cdot \frac{[Al]^2}{[BC]^3} \tag{101}$$

where frH_{ac}, $frBC_{ac}$, and $frAl_{ac}$ are the equivalent fractions of H, BC and Al on the adsorption complex and KH_{ex} and KAl_{ex} are the Gaines-Thomas selectivity constants for H/BC exchange and Al/BC exchange, respectively. The description of the exchange between H and Al is obtained by combining the equations (100) and (101). Since the exchange complex is assumed to comprise H, Al and BC only, charge balance requires that

$$frH_{ac} + frAl_{ac} + frBC_{ac} = 1 \tag{102}$$

Sulphate adsorption is described by a Langmuir equation according to:

$$SO_{4,ac} = \frac{SSC \cdot kSO_{4,ad} \cdot [SO_4]}{1 + kSO_{4,ad} \cdot [SO_4]} \tag{103}$$

where $SO_{4,ac}$ is the sulphate content at the adsorption complex ($mol_c\ kg^{-1}$), SSC is the sulphate sorption capacity ($mol_c\ kg^{-1}$) and $kSO_{4,ad}$ is the sulphate adsorption constant ($m^3\ mol_c^{-1}$).

A complete description of SMART including the mathematical procedure for solving the set of mass balance and equilibrium equations as well as the initialization procedure is described in de Vries et al. (1989a). However, sulphate adsorption and dynamic N immobilization is not described there, as this is only recently included in the model.

3.6 The dynamic multi-layer model RESAM

3.6.1 Introduction

The dynamic approach described before can also be combined with a nutrient cycle. Such a model has been developed for the Netherlands (de Vries en Kros, 1990) as part of the Dutch Acidification and Simulation model DAS. This soil model called RESAM (Regional Soil Acidification Model) is by far more complicated than SMART. As with MACAL, the various base cations are not lumped and RESAM includes the same descriptions for canopy interactions and litterfall. Additional processes which are neither described in SMART or MACAL are root decay, mineralization and complexation reactions with organic anions. Furthermore, most processes are described by first order reactions and not by equilibrium equations (e.g. carbonate weathering and hydroxide weathering) and cation exchange comprises seven cations. Moveover, unlike SMART, RESAM is a multi-layer model. RESAM has been developed specifically for application on a national scale. Such applications have already been made (De Vries et al., 1991a).

As with all former models, RESAM assumes that:
- immobilization, reduction and precipitation of SO_4 is negligible;
- nitrogen fixation is negligible.

Analogous to SMART, it is furthermore assumed that:
- all soil layers are homogeneous compartments of constant density;
- the element input mixes completely in all soil layers; and
- the water flow is stationary which implies that the water flux percolating from a soil layer equals the infiltration minus the transpiration.

3.6.2 Basic principle

RESAM consists of a set of mass balance equations, equilibrium equations and rate-limited equations. An overview of the processes in RESAM in relation to the various ions considered is given in Table 3.

Analogous to SMART, the mass balance equations describe the input-output relationships for all cations and anions (except for HCO_3 and H) in each soil layer i according to:

$$\frac{dX_{tot(i)}}{dt} = X_{in}(i-1) + X_{int}(i) - FW(i) \cdot [X](i) \qquad (104)$$

The model contains a mass balance for Ca in carbonates, for Ca, Mg, K and Na in litter, primary minerals and at the adsorption complex, for Al in hydroxides and at the adsorption complex, for N in litter and in needles and for S in litter and at the adsorption complex.

Table 3 Overview of the ions and processes included in RESAM. ("+" = ion included, "-" = ion not included)

Process	H	Al	Ca	Mg	K	Na	NH_4	NO_3	SO_4	Cl	HCO_3	RCOO
Deposition[1)]	+	-	+	+	+	+	+	+	+	+	-	-
Foliar uptake[1)]	+	-	-	-	-	-	+	+	+	-	-	-
Foliar exudation[1)]	+	-	+	+	+	-	-	-	-	-	-	-
Litterfall[1)]	-	-	+	+	+	-	+	+	+	-	-	-
Root decay[5)]	-	-	+	+	+	-	+	+	+	-	-	-
Mineralization[5)]	+	-	+	+	+	-	+	+	+	-	-	+
Maintenance uptake[2)]	+	-	+	+	+	-	+	+	+	-	-	-
Growth uptake[2)]	+	-	+	+	+	-	+	+	+	-	-	-
Nitrification[4)]	+	-	-	-	-	-	+	+	-	-	-	-
Denitrification[4)]	+	-	-	-	-	-	-	+	-	-	-	-
Dissociation[2)]	+	-	-	-	-	-	-	-	-	-	+	-
Protonation[5)]	+	-	-	-	-	-	-	-	-	-	-	+
Carbonate weathering[4)]	+	-	+	-	-	-	-	-	-	-	+	-
Silicate weathering[4)]	+	+	+	+	+	+	-	-	-	-	-	-
Al-hydroxide weathering[4)]	+	+	-	-	-	-	-	-	-	-	-	-
Cation exchange[3)]	+	+	+	+	+	+	+	-	-	-	-	-
Sulphate adsorption[3)]	+	-	-	-	-	-	-	-	+	-	-	-

1) These input terms are described exactly similar as in MACAL.
2) These interaction terms are described (nearly) similar as in MACAL.
3) These interaction terms are described (nearly) similar as in SMART.
4) These interaction terms are described different from MACAL and SMART.
5) These interaction terms are not described in MACAL and SMART.

The concentration of HCO_3 in each layer is calculated with equation (40) using a constant CO_2 pressure. The H concentration is determined by the charge balance equation:

$$[H]+[Al]+[Ca]+[Mg]+[K]+[Na]+[NH_4] = [NO_3]+[SO_4]+[Cl]+[HCO_3]+[RCOO] \qquad (105)$$

3.6.3 Input and interaction fluxes

Unlike MACAL, RESAM contains a litter layer and an explicit description for the mineralization in this layer. The input fluxes to this layer are equal to the input by throughfall. This is equal to the total deposition corrected for foliar exudation (Ca, Mg and K) and for foliar uptake (SO_4, NH_4 and NO_3). Unlike MACAL, litterfall is not included as an input flux to the soil solution (cf equation (66) to (69)) but to an organic pool. As with MACAL, foliar uptake, foliar exudation and litterfall is discribed according to equation (70) to (78).

The various interaction fluxes in RESAM in combination with the relevant ions are listed in Table 3. A description of these processes is given below. Except for cation exchange and sulphate adsorption, all processes have been described by rate-limited (mostly first-order) reactions.

Root decay

Input of N, Ca, Mg, K and S by root decay is described similar as litterfall, i.e.:

$$X_{rd} = (1 - fr_{re})\, k_{rd} \cdot Am_{rt} \cdot ctX_{rt} \tag{106}$$

where X_{rd} is the flux of element X due to root decay ($mol_c\ ha^{-1}\ yr^{-1}$), k_{rd} is the root decay constant (yr^{-1}), Am_{rt} is the amount of fine roots ($kg\ ha^{-1}$) and ctX_{rt} is the content of element X in fine roots ($mol_c\ kg^{-1}$).

As with litterfall, reallocation is limited to N (cf equation (76) and (77)). The reallocationfraction, fr_{re} is described as a function of the N content in roots similar to equation (78). Root decay is limited to fine roots which appear to be very dynamic. The occurence of this process with depth is determined by the distribution of fine roots.

Mineralization

For the simulation of the decomposition of above ground organic matter (litter) a distinction is made between a rapidly decomposing pool of fresh litter (less than 1 year) and a slowly decomposing pool of old litter (more than 1 year) (Janssen, 1984).

The mineralization of N, Ca, Mg, K and S from fresh litter is described as:

$$X_{mi,lf} = (fr_{le} + fr_{mi} \cdot (1 - fr_{le})) \cdot X_{lf} \tag{107}$$

where fr_{mi} is a mineralization fraction (-) and fr_{le} is a leaching fraction (-).

Leaching only refers to the release of cations from fresh litter just after litterfall. It is a process which is especially important for K. For this cation the leaching fraction is very high. Consequently, the K content in litter is extremely low. Actually, leaching is a process which differs from mineralization since organic matter is not decomposed. However, both processes have been lumped since both leaching and mineralization lead to a release of elements to the soil solution.

During mineralization, nitrogen is released as NH_4. Fresh litter which is not mineralized is transferred to the old litter (humus) pool. The mineralization of N, Ca, Mg, K and S from this pool is described by first order kinetics (Van Veen, 1977):

$$X_{mi,lt} = k_{mi,lt} \cdot Am_{lt} \cdot ctX_{lt} \tag{108}$$

where $k_{mi,lt}$ is the mineralization rate constant from litter (yr^{-1}), Am_{lt} is the amount of old litter ($kg\ ha^{-1}$) and ctX_{lt} is the content of element X in litter ($mol_c\ kg^{-1}$).

At present, mineralization of organic matter in the mineral soil layers is not considered in RESAM, except for the mineralization from dead fine roots, which are fed by root decay as described before. As with old litter, mineralization of dead roots is described as:

$$X_{mi,dr} = k_{mi,dr} \cdot Am_{dr} \cdot ctX_{dr} \tag{109}$$

where $k_{mi,dr}$ is the mineralization rate constant from dead roots or root necromass (yr^{-1}), Am_{dr} is the amount of dead roots ($kg\ ha^{-1}$) and ctX_{dr} is the content of element X in dead roots ($mol_c\ kg^{-1}$).

Rate constants (and fractions) describing biochemical processes (mineralization, nitrification and denitrification) are described in RESAM as maximum values, which are reduced for environmental factors such as soil moisture (groundwater level) and pH. The mineralization fraction , fr_{mi}, and mineralization rate constants, $k_{mi,lt}$ and $k_{mi,dr}$, are reduced with decreasing mean groundwater levels. For nitrogen, the values are also reduced at low N contents (high C/N ratios) to account for immobilization in microbes according to (Janssen, 1983):

$$fNred_{mi} = 1 + \frac{1}{DAR} \cdot \left(\frac{C/N_{mo} - C/N_s}{C/N_{mo}}\right) \tag{110}$$

where $fNred_{mi}$ is the reduction fractor by which the mineralization fraction and rate constants have to be multiplied (-), C/N_{mo} is the C/N ratio of the micro-organism decomposing the substrate (-), C/N_s is the C/N ratio of the substrate (fresh litter, old litter or dead roots) and DAR is the dissimilation to assimilation ratio of the decomposing micro-organisms (-). Values for DAR and C/N_{mo} are related to funghi because they are mainly responsible for mineralization of forest litter.

It should be noted that equation (110) only holds for $C/N_{mo} < C/N_s < (1+DAR) \cdot C/N_{mo}$. When C/N_s is less then C/N_{mo}, $fNred_{mi} = 1$ and when C/N_{mo} is more than $(1+DAR) \cdot C/N_{mo}$, $fNred_{mi} = 0$.

Organic anions, which are also produced during mineralization, are calculated in RESAM from charge balance considerations:

$$RCOO_{mi} = NH_{4,mi} + Ca_{mi} + Mg_{mi} + K_{mi} - SO_{4,mi} \tag{111}$$

where *mi* stands for the mineralization from both fresh and old litter and from dead roots.

Root uptake

As with MACAL, total root uptake of Ca, Mg, K, S and N is described as a demand function, which consists of maintenance uptake, to resupply the needles/leaves and roots, and net (growth) uptake in stems and branches. Total uptake fluxes are equal to (cf equations (85), (86) and (89)):

$$X_{ru} = X_{lf} + X_{rd} + X_{fe} + X_{gu} \tag{112}$$

$$S_{ru} = S_{lf} + S_{rd} - S_{fu} \tag{113}$$

$$N_{ru} = N_{lf} + N_{rd} - NH_{4,fu} + N_{gu} \tag{114}$$

with X is Ca, Mg or K. For growth uptake, RESAM has two options, a constant growth or a logistic growth.

The uptake from a given soil layer is derived by multiplying the total uptake flux with the ratio of transpiration (water uptake) from that layer to the total transpiration. As with MACAL, nutrient and water uptake are thus coupled in RESAM. Unlike START, SMART and MACAL, RESAM includes the possibility for preferent uptake of NH_4.

The uptake of NH_4 and NO_3 is calculated as (cf equation (87) and (88)):

$$NH_{4,ru} = fNH_{4,ru} \cdot \left(\frac{[NH_4]}{[NH_4]+[NO_3]}\right) \cdot N_{ru} \tag{115}$$

$$NO_{3,ru} = N_{ru} - NH_{4,ru} \tag{116}$$

where $fNH_{4,ru}$ is a preference factor for the uptake of NH_4 above NO_3. When $fNH_{4,ru}$ equals 1, the preference for the uptake of NH_4 and NO_3 is equal. The inclusion of the option of preferent NH_4 uptake is based on literature information (e.g. Gijsman, 1990). When $NH_{4,ru}$ exceeds N_{ru} (cf equation (15)), NH_4 uptake is set equal to N uptake. This implies that all N is taken up as NH_4.

Nitrification and denitrification

Both nitrification and denitrification are described in RESAM as first order reactions according to:

$$NH_{4,ni} = - \theta \cdot D \cdot k_{ni} \cdot [NH_4] \tag{117}$$

$$NO_{3,de} = - \theta \cdot D \cdot k_{de} \cdot [NO_3] \tag{118}$$

where θ is the volumetric water content ($m^3\ m^{-3}$), D is the layer thickness (m), k_{ni} and k_{de} are the nitrification and denitrification rate constant (yr^{-1}) respectively.

As with mineralization, the maximum values for the nitrification and denitrification rate constant are affected by the mean groundwater level. Values are reduced with a decreasing mean groundwater level for nitrification since this process is restricted to aerobic conditions. For denitrification, the opposite is true. Both rate contents are also reduced with decreasing pH. A description of the reduction functions is given in De Vries et al. (1988).

The nitrification rate constant is also reduced according to the presence of organic matter. Consequently, the nitrification rate is high in the litter layer whereas it reduces to zero in the subsoil. This is in accordance with field data (Tietema and Verstraten, 1988; Tietema et al., 1990; De Boer, 1989). Finally, the nitrification rate constant in the litter layer is reduced with an increase in thickness of this layer. This is based on various literature sources (Lensi et al., 1986; Clays-Josserand et al., 1988; Tietema, et al., in prep.) and can be explained by the fact that litter forms a barrier for O_2 diffusion from the atmosphere, thus creating unfavourable anaerobic conditions for nitrifying bacteria (Lensi et al., 1986). The description is such that above a thickness of 5 cm, the product of D and k_{ni} (cf equation (117)) does not increase any more.

Protonation

Protonation refers to the association of organic anions with the hydrogen ion. Protonation is in fact an equilibrium process according to:

$$RCOO^- + H^+ \rightleftharpoons RCOOH$$

At low pH, the equilibrium is forced to the right hand side. In RESAM, protonation is provisionally described by a first order reaction, with a pH dependent rate constant to account for the equilibrium affect, according to:

$$RCOO_{pr} = -\ \theta \cdot D \cdot k_{pr} \cdot [RCOO] \tag{119}$$

where k_{pr} is the protonation rate constant (yr^{-1}).

Weathering

As with SMART, three types of mineral pools are distinghuished in RESAM, i.e. carbonates, silicates (primary minerals) and aluminium oxides. However, unlike SMART, the dissolution of Al and base cations from all these minerals is described by rate limited expressions using first order kinetics according to:

$$Ca_{we,cb} = \rho \cdot D \cdot kCa_{we,cb} \cdot ctCa_{cb} \cdot ([Ca_e] - [Ca]) \tag{120}$$

$$X_{we,pm} = \rho \cdot D \cdot kX_{we,pm} \cdot ctX_{pm}\ \ [H]^{\alpha} \tag{121}$$

$$Al_{we,ox} = \rho \cdot D \cdot kAl_{we,ox} \cdot ctAl_{ox} \cdot ([Al_e] - [Al]) \tag{122}$$

where ρ is the bulk density ($kg\ m^{-3}$); $kCa_{we,cb}$, $kX_{we,pm}$ and $kAl_{we,ox}$ are the weathering rate constants ($m^3\ mol_c^{-1}\ yr^{-1}$) of Ca from carbonates of base cations from primary minerals and of Al from hydroxides respectively; $ctCa_{cb}$, ctX_{pm} and $ctAl_{ox}$ are the contents ($mol_c\ kg^{-1}$) of Ca in carbonates, base cations (Ca, Mg, K and Na) in primary minerals and Al in hydroxides respectively; α is a dimensionless exponent; $[Ca_e]$ and $[Al]_e$ are the Ca and Al concentratiorı ($mol_c\ m^{-3}$) in equilibrium with calcite and gibbsite respectively and [Ca], [H] and [Al] are the actual concentrations ($mol_c\ m^{-3}$) of Ca, H and Al respectively.

The rate limited expression for carbonate weathering is based on the generally measured exponential decrease of calcium carbonate with time. This can be interpreted as the result of increasingly incomplete contact of the percolating water with larger shell fragments, that remain when decalcification proceeds (Klijn, 1981). When the soil solution is supersaturated with respect to calcite, equilibrium is enforced. The equilibrium concentration is calculated according to equation (99).

The kinetic expression for base cation weathering is based on laboratory experiments (Van Grinsven et al., 1988b). The influence of pH on the weathering rate (cf equation (121)) is particularly important for Mg and K weathering in the very acid range (Van Grinsven et al., 1988b). At present, the value of α is set to zero, by which the weathering rate is made independent of pH. The production of Al from the congruent dissolution of silicate minerals is directly related to the production of base cations. Standard minerals are assumed as pools for K (Microcline), Na (Albite), Ca (Anorthite) and Mg (Chlorite). Stoichiometric ratios of Al to base cations ($mol_c\ mol_c^{-1}$) in these minerals are 3 for K, 3 for Na, 3 for Ca and 0.6 for Mg. Microline and albite are commonly present in the sandy soils in the Netherlands, but the assumption that Ca originates from anorthite and Mg from chlorite is poorly supported by field data. There are various minerals containing Ca and Mg but their nature and contribution to the release of Ca and Mg is difficult to determine.

The rate limited expression for Al-hydroxide weathering is based on the generally measured equilibrium with gibbsite in sandy subsoils, whereas undersaturation occurs in the topsoil (e.g. Cronan et al., 1986: Mulder and Van Breemen, 1987). In the model, $ctAl_{ox}$ is restricted to the amount of amorphous hydroxides, since gibbsite (a cristalline phase) rarely occurs in acid sandy soils (cf section 2.4). As with calcite, equilibrium is enforced with respect to Al hydroxide when the soil solution is supersaturated. The equilibrium concentration is calculated according to equation (39).

Regarding Al weathering, RESAM also contains the option of an Elovich-equation according to:

$$Al_{we,ox} = \rho \cdot D \cdot kEL_{o,1} \cdot \exp\,(kEL_{o,2} \cdot ctAl_{ox}) \cdot ([Al_e] - [Al]) \tag{123}$$

where $kEL_{o,1}$ ($m^3\ kg^{-1}\ yr^{-1}$) and $kEL_{o,2}$ ($kg\ mol_c^{-1}$) are the so called Elovich constants.

Use of this equation, which is also used in literature for the description of P adsorption (e.g. Van Riemsdijk and De Haan, 1981) is based on laboratory experiments on acid neutralization by Al mobilization (Van Grinsven et al., 1988a; De Vries et al., in prep.).

Adsorption and desorption

Adsorption and desorption reactions are included in RESAM as equilibrium equations for all cations involved and for SO_4. The adsorption flux is calculated as:

$$X_{ad} = \rho \cdot D \cdot (ctX_{ac(t)} - ctX_{ac(t-1)}) \tag{124}$$

where ctX_{ac} is the content of element X (H, Al, Ca, Mg, K, Na, NH_4 and SO_4) on the adsorption complex ($mol_c\ kg^{-1}$), t refers to the current and t-1 to the preceding timestep.

As with SMART, cation exchange is described by Gaines-Thomas equations with Ca as a reference ion according to (cf equation (100) and (101)):

$$\frac{(frX_{ac})^2}{(frCa_{ac})^{z}x} = KX_{ex} \cdot \frac{[X]^2}{[Ca]^{z}x} \tag{125}$$

where z_x is the valence of cation x, KX_{ex} is the Gaines-Thomas selectivity constant for exchange of cation X against Ca and frX_{ac} is the equivalent fraction of cation X on the adsorption complex (-) which is calculated as:

$$frX_{ac} = ctX_{ac} \ / \ CEC \tag{126}$$

where CEC is the cation exchange capacity ($mmol_c \ kg^{-1}$).

In order to solve equation (125) for six ions (H, Al, Mg, K, Na and NH_4) with seven unknowns (the adsorbed fractions of the same ions plus Ca) an additional requirement is met (cf equation (102)):

$$frH_{ac} + frAl_{ac} + frCa_{ac} + frMg_{ac} + frK_{ac} + frNa_{ac} + frNH_{4,ac} = 1 \tag{127}$$

As with SMART, SO_4 adsorption in each soil layer is described with a Langmuir (equilibrium) equation (equation (103)).

3.7 Summary

A summarizing overview of the processes and process formulations included in the various steady state and dynamic models is given in Table 4.

Table 4 Processes and process formulations included in START, MACAL, SMART and RESAM

Processes	START	MACAL	SMART	RESAM
Hydrological processes:				
Water flow	Precipitation surplus	Variable flow with depth	Precipitation surplus	Hydrologic submodel
Biogeochemical processes:				
Foliar uptake	-	Proportional to total deposition	-	Proportional to total deposition
Foliar exudation	-	Proportional to H and NH_4 deposition	- -	Proportional to H and NH_4 deposition
Litterfall	-	First order reaction	-	First order reaction
Root decay	-	-	-	First order reaction
Mineralization/ immobilization	Zero order reaction[1)]	Zero order reaction[1)]	Proportional to N deposition	First order reaction
Growth uptake	Constant growth	Constant growth	Constant growth	- Constant growth - Logistic growth
Maintenance uptake		-	Forcing function[2)]	-Forcing function[2)]
Nitrification	Proportional to net NH_4 input	Proportional to NH_4 flux	Proportional to net NH_4 input	First order reaction
Denitrification	Proportional to net NO_3input	Proportional to NO_3 flux	Proportional to net NO_3 input	First order reaction
Geochemical processes:				
CO_2 dissociation	Equilibrium	Equilibrium	Equilibrium	Equilibrium
RCOO protonation	-	-	-	First order reaction
Carbonate weathering	-	-	Equilibrium	First order reaction
Silicate weathering	Zero order reaction	Zero order reaction	Zero order reaction	First order reaction[3)]
Al-hydroxide weathering	Gibbsite equilibrium	Gibbsite equilibrium	Gibbsite equilibrium	- First order reaction - Elovich equation
Cation exchange	-	-	Gaines Thomas equation[4)]	Gaines Thomas equation[4)]
Sulphate adsorption	-	-	Langmuir equation	Langmuir equation

1) START and MACAL only include long-term net nitrogen immobilization.
2) In MACAL the maintenance uptake equals litterfall plus foliar exudation minus foliar uptake and in RESAM it equals the sum of litterfall, root decay and foliar exudation minus foliar uptake.
3) In RESAM there is also the option to include a dependence of pH on the weathering rate.
4) In SMART cation exchange is limited to H, Al and BC whereas it includes H, Al, Ca, Mg, K, Na, NH_4 in RESAM.

4 RECEPTOR AREAS AND RECEPTOR DISTRIBUTION DISTRIBUTION

4.1 Receptor areas

Emissions and thereby deposition fluxes strongly vary in space. Consequently, receptor - or deposition areas have to be defined, to attain an optimum between the number of areas and the spatial variability within each area. The shape and spatial resolution of a receptor area directly affects the mapping procedure.

Maps can either be displayed as polygons or as grids. The advantage of polygon maps is that so called functional subregions can be displayed with a similar critical load. In this context, functional subregions can be defined as geographic areas with similar environmental characteristics (e.g. soil type, elevation class etc.). characterizing receptor response. However, the disadvantage is the lack of uniformity in geographic presentation. Consequently, a grid system is used both for the European and National application.

Two types of grid systems which are commonly used today in Europe are:

1 The EMEP grid system of 150 x 150 km, used for modelling deposition of SO_2, NO_x and NH_3 for given emission rates.

2 The RAINS grid system of 1.0° longitude x 0.5° latitude (approximately 50 x 50 km), used for modelling environmental impacts for given deposition rates.

For the European application of the START and SMART model the RAINS grid is used. Using this 1.0° x 0.5° grid the length of a grid element in the south-north direction is fixed to 56 km but the width in the east-west direction varies between 38 to 91 km depending on the latitude (approximately 50 to 60 km in Central Europe). The total number of grids equals 2364. For the Netherlands, where much more detailed information regarding atmospheric deposition soils and vegetation does exist, a 10 x 10 km grid is used for the assessment of critical loads. The number of grids containing forests equals 434. However, for the assessment of long-term impacts on Dutch forest soils, with the RESAM model, use is made of 20 receptor areas with relevant statistical information on emissions (De Vries, 1990).

4.2 Receptors and receptor distribution

4.2.1 European application

The receptors that are of interest in this report are forest ecosystems, i.e. combinations of tree species and soil type (cf section 1.2). Both for the assessment of critical loads (with the START model) and for the evaluation of long-term impacts (with the SMART model), on European forest soils the same receptors are considered. Regarding tree species, a distinction is made between coniferous and deciduous trees. Although detailed information on the areal distribution of various tree species (Pine, Fir, Spruce, Oak, Beech and Birch) is available, this has not been used since the spatial allocation of individual tree species over soil types was not known.

Soil types are distinguished on the basis of the FAO-UNESCO Soil Map of the World (FAO-UNESCO, 1981). European soils on the FAO-UNESCO map have been classified into 80 soil types given in Table 5.

Table 5 Soil units considered for Europe (in alphabetical order).

A	ACRISOLS	I	LITHOSOLS	R	REGOSOLS
Ao	Orthic acrisols			Re	Eutric regosols
Af	Ferric acrisols	J	FLUVISOLS	Rc	Calcaric regosols
Ah	Humic acrisols	Je	Eutric fluvisols	Rd	Dystric regosols
		Jc	Calcaric fluvisols	Rx	Gelic Regosols
B	CAMBISOLS	Jd	Dystric fluvisols		
Be	Eutric cambisols	Jt	Thionic fluvisols	S	SOLONETZ
Bd	Dystric cambisols			So	Orthic solonetz
Bh	Humic cambisols	K	KASTANOZEMS	Sm	Mollic solonetz
Bg	Gleyic cambisols	Kh	Haplic kastanozems	Sg	Gleyic solonetz
Bk	Calcic cambisols	Kk	Calcic kastanozems		
Bc	Chromic cambisols	Kl	Luvic kastanozems	T	ANDOSOLS
Bv	Vertic cambisols			To	Orthic andosols
		L	LUVISOLS	Tm	Mollic andosols
C	CHERNOZEMS	Lo	Orthic luvisols	Th	Humic andosols
Cg	Glossic chernozems	Lc	Chromic luvisols	Tv	Vitric andosols
Ch	Haplic chernozems	Lk	Calcic luvisols		
Ck	Calcic chernozems	Lv	Vertic luvisols	U	RANKERS
Cl	Luvic chernozems	Lf	Ferric luvisols		
		La	Albic luvisols	V	VERTISOLS
D	PODZOLUVISOLS	Lg	Gleyic luvisols	Vp	Pellic vertisols
De	Eutric podzoluvisols			Vc	Chromic vertisols
Dd	Dystric podzoluvisols	M	GREYZEMS		
Dg	Gleyic podzoluvisols	Mo	Orthic greyzems	W	PLANOSOLS
				We	Eutric planosols
E	RENDZINAS	O	HISTOSOLS	Wd	Dystric planosols
		Oe	Eutric histosols		
G	GLEYSOLS	Od	Dystric histosols	X	XEROSOLS
Ge	Eutric gleysols	Ox	Gelic histosols	Xh	Haplic xerosols
Gc	Calcaric gleysols			Xk	Calcic xerosols
Gd	Dystric gleysols	P	PODZOLS	Xy	Gypsic xerosols
Gm	Mollic gleysols	Po	Orthic podzols	Xl	Luvic xerosols
Gh	Humic gleysols	Pl	Leptic podzols		
Gx	Gelic gleysols	Ph	Humic podzols	Y	YERMOSOLS
		Pp	Placic podzols	Yk	Calcic yermosols
H	PHAEOZEMS	Pg	Gleyic podzols		
Hh	Haplic phaeozems			Z	SOLONCHAKS
Hc	Calcaric phaeozems	Q	ARENOSOLS	Zo	Orthic solonchaks
Hl	Luvic phaeozems	Qc	Cambic arenosols	Zm	Mollic solonchaks
Hg	Gleyic phaeozems	Ql	Luvic arenosols	Zg	Gleyic solonchaks

The number of the mapping units on the 1 : 5 000 000 FAO-UNESCO map is, however more than 300 since the code of a mapping unit contains four items. i.e.
- the dominant soil unit : one or two letter symbol, e.g. Po
- the associated soil unit : one or two digit symbol, e.g. 25
- the dominant texture class: one digit symbol, e.g. 2
- the dominant slope class : one letter symbol, e.g. a

Mapping units on the FAO soil map of Europe only occasionally consist of one soil unit. Generally, it is an association of a dominant soil unit, associated soils which also cover at least 20% of the mapping unit and/or inclusions covering less than 20%.

For this application the soils are distinguished on the basis of the dominant soil unit, texture class and slope class. Texture classes are defined as:
1 coarse : clay content less than 18%
2 medium : clay content between 18 and 35%
3 fine : clay content above 35%
When two texture classes occur within one mapping unit, this is indicated as 1/2, 2/3 or 1/3.

Slope classes have been defined as:
a even : slope less than 8%
b undulating : slope between 8 and 30%
c steep : slope above 30%
Similar to texture classes, several slope classes may occur within one mapping unit, indicated as ab or bc.

The areal distribution of soils was digitized by estimating the fraction of each mapping unit (soil type, texture class and slope class) within each grid square using the FAO-UNESCO soil map. The resolution of this map is such that a grid contain one to seven mapping units, the mean number being 2.2. As with soils, the areal distribution of forests has been digitized by estimating the fraction of coniferous and deciduous forests in each grid square using aeronautic maps.

The distributions of both soils and forests within a grid is not known. In estimating the spatial distribution of forest soils, it has been assumed that forests are not evenly distributed over all soil types, but instead, they are mainly located on areas with steep slopes and poor soils (low weathering rates and coarse texture). The fraction of forest-soil combinations in each grid was estimated using the following allocation procedure: first, forests were allocated on soils with steep slopes (slope classes c and bc) followed by non-calcareous coarse textured soils, peat soils, calcareous coarse textured soils, non-calcareous medium and fine textured soils and calcareous medium and fine textured soils.

In Europe 28.5% of the total land area consists of forests. About 65% are coniferous forests and 35% deciduous forests. Forest percentages in the various countries vary from less than 1% in Ireland to more than 60% in Finland.

An overview of the most important forest soils, covering more than 1% of the forested area in Europe, is given in Table 6.

Table 6 Area of the most important forest soils in Europe, as a percentage of the total forest area

Soil code	Soil type (FAO)	Area (%)	
		uneven distribution	even distribution
Po	Orthic Podzol	33.8	31.4
De	Eutric Podzoluvisol	12.6	12.7
Bd	Dystric Cambisol	9.4	8.6
Be	Eutric Cambisol	6.4	5.8
Lg	Gleyic Luvisol	4.9	4.5
Lo	Orthic Luvisol	3.6	4.1
Od	Dystric Histosol	3.5	4.5
Dd	Dystric Podzoluvisol	3.3	3.6
Bk	Calcic Cambisol	2.7	3.1
E	Rendzina	2.5	2.9
Pl	Leptic Podzol	2.2	1.5
Lc	Chromic Luvisol	1.4	2.1
Pg	Gleyic Podzol	1.2	1.1
I-U	Lithosol-Ranker	1.1	0.5
Bh	Humic Cambisol	1.1	0.9

Table 6 also gives information on the forest coverage of several soil types using an even distribution of forests over the soil types in a grid. The difference with the forest allocation described above appears to be very small.

Most forests are located on podzols and podzoluvisols, especially in the Nordic countries, and to a lesser extent on cambisols and luvisols. More than 80% of the forests are located on these soil types, i.e. nearly 40% on podzols, more than 15% on podzoluvisols and cambisols and about 10% on luvisols. The occurence of forest on all other soil types appears to be less than 5% (cf Table 6).

4.2.2 Application in the Netherlands

For the assessment of critical loads on a national scale, a distinction has been made in 12 tree species and 23 soil types. The total forested area in the Netherlands is about 320 000 ha, which is approximately 9.5% of the total area of the Netherlands. Tree species included are Pinus Sylvestris (Scotch Pine; 38.2%), Pinus Nigra (Black Pine; 5,9%), Pseudotsuga Menziesii (Douglas Fir; 5.5%), Picea Abies (Norway Spruce; 5.1%), Larix Leptolepis (Japanese Larch; 5.7%), Quercus Robur (Oak; 17.4%), Fagus Sylvatica (Beech; 4.1%), Populus Spec (Poplar; 4.6%), Salix Spec (Willow; 2.4%), Betula Pendula (Birch; 7.4%), Fraxinus Nigra (Ash; 1.9%) and Alnus Glutinosa (Black Alder; 1.9%). Soil types were differentiated in 18 non-calcareous sandy (mainly podzolic) soils (84.6%), calcareous sandy soils (2.2%), loess soils (1.5%), non-calcareous clay soils (4.0%), calcareous clay soils (4.2%) and peat soils (3.5%) on the basis of a recent 1 : 250 000 soil map of the Netherlands (De Vries et al., 1991b).

Information on the area (distribution) of each specific forest-soil combination in a grid was derived by overlaying the digitized forest- and soil data base. This was done by a grid-overlay of the digitized 1 : 250 000 soil map with a spatial resolution of 100 m x 100 m and a data base with tree species information with a spatial resolution of 500 x 500 m for each 10 x 10 km grid (De Vries et al., 1991b). An overview of the area of the 18 non-calcareous sandy soils included, which cover about 85% of the Dutch forest, area is given in Table 7. The total number of forest-soil combinations for all grids equals 17102, i.e. 12514 on non-calcareous sandy soils and 4588 on all other soils. The number of forest/soil combinations in a grid varies between 1 and 125.

For the assessment of long term impacts on forest soils in the Netherlands (with the RESAM model) the receptors are restricted to tree species and soil types of major importance to limit the computation time. Tree species includes are: Pinus Sylvestris (Scotch Pine), Pinus Nigra (Black Pine), Pseudotsuga Menziesii (Douglas Fir), Picea Abies (Norway Spruce), Larix Leptolepis (Japanese Larch), Quercus Robur (Oak) and Fagus Sylvatica (Beech). Forest soils are confined to acid sandy soils only. These soils cover a large forest area and are sensitive to acidification. The soils are characterized by 14 representative soil types indicated in Table 7. These soil types cover a broad range in soil properties, such as the organic matter content, texture and groundwater level, which influence major hydrological, biochemical and geochemical processes in the soil. The vertical heterogeneity is taken into account by differentiating between soil layers (horizons). An overview of the designation and thickness of the horizons in the various soil profiles is given in De Visser and De Vries (1989).

The forest/soil combinations that have been included cover nearly 65% of the total Dutch forest area (cf Table 7) of which more than 50% is covered by Pinus Sylvestris (Scotch Pine). The remaining 35% comprises tree species such as Populus Spec (Poplar) and Betula Pendula (Birch) (approximately 20%) and soil types such as calcareous sandy soils, clay soils, loess soils and peat soils (approximately 15%). The spatial distribution (area) of the forest/soil combinations considered in each of the 20 receptor areas has also been assessed by a grid-overlay procedure.

Table 7 Area of the non-calcareous sandy forest soils in the Netherlands as a percentage of the total forest area

Soil code	Soil type: Netherlands[1]	USDA[2]	FAO[3]	Area (%)
Z5	Holtpodzol, fijnzandig zeer diep ontwaterd	Entic Haplorthod	Leptic Podzol fine textured	2.7
Z6	Holtpodzol, grofzandig zeer diep ontwaterd	Entic Haplorthod	Leptic Podzol coarse textured	8.7
Z7[4], Z8	Veldpodzol, fijnzandig matig ontwaterd	Typic Haplorthod	Gleyic Podzol fine textured	14.9
Z9[4]	Veldpodzol, grofzandig matig ontwaterd	Typic Haplothod	Gleyic Podzol coarse textured	0.3
Z11[4]	Laarpodzol, fijnzandig diep ontwaterd	Plaggeptic Haplaquod	n.s.e.[5]	-
Z12	Haarpodzol, fijnzandig zeer diep ontwaterd	Typic Haplohumod	Humic Podzol fine textured	11.3
Z13	Haarpodzol, grofzandig zeer diep ontwaterd	Typic Haplohumod	Humic Podzol coarse textured	8.3
Z14[4], Z16	Enkeerd, fijnzandig diep ontwaterd	Plaggept	Anthrosol	3.1
Z15[4]	Enkeerd, grofzandig diep ontwaterd	Plaggept	Anthrosol	0.1
Z17[4], Z18	Loopodzol, fijnzandig ondiep/diep ontwaterd	n.s.e.[5]	n.s.e.[5]	4.4
Z19[4]	Loopodzol, grofzandig ondiep/diep ontwaterd	n.s.e.[5]	n.s.e.[5]	0.6
Z20	Beekeerd, fijnzandig zeer ondiep ontwaterd	Typic Humaquept	Humic Gleysol fine textured	2.9
Z21	Gooreerd, fijnzandig ondiep ontwaterd	Typic Humaquept	Humic Gleysol fine textured	1.2
Z22[4]	Gooreerd, grofzandig ondiep/diep ontwaterd	Typic Humaquept	Humic Gleysol coarse textured	0.1
Z23, Z24[4]	Vlakvaag, fijnzandig ondiep ontwaterd	Typic Psammaquent	Fluvisol fine textured	0.5
Z25[4]	Vlakvaag, grofzandig ondiep/diep ontwaterd	Typic Psammaquent	Fluvisol coarse textured	0.0
Z27	Duinvaag, fijnzandig zeer diep ontwaterd	Typic Udipsamment	Albic Arenosol fine textured	13.1
Z28	Duinvaag, grofzandig zeer diep ontwaterd	Typic Udipsamment	Albic Arenosol coarse textured	1.0
-	Associations	-	-	11.4

1) Soil map of the Netherlands (1985)
2) Soil Survey Staff (1975)
3) FAO (1988)
4) Not included in the RESAM applications
5) No suitable equivalent

5 DATA

Data that are needed by the various models can be divided in inputs and outputs to the soil system and soil data. In the following sections an overview is given of the acquisition strategy for both types of data including values that will be used in regional applications.

5.1 Inputs and outputs to the soil system

5.1.1 Data acquisition strategy

Inputs to the soil system are atmospheric deposition of SO_4, NO_3, NH_4, Cl and base cations, corrected for foliar uptake (SO_4, NO_3, NH_4) and foliar exudation (Ca, Mg, K) in the forest canopy, nutrient (N, Ca, Mg, K, S) input by litterfall and root decay and precipitation. Systems outputs include nutrient uptake and evapotranspiration.

The inputs and outputs mentioned above vary as a function of location (receptor area) and receptor (the combination of tree species and soil type) as shown in Table 8.

Table 8 The influence of location, tree species and soil type on inputs and outputs to the soil system

Inputs/Outputs	Location	Tree species	Soil type
Atmospheric deposition	x	x	-
Canopy interactions[1)]	x	x	-
Litterfall and root decay[1)]	-	x	-
Uptake	x[2)]	x	x[3)]
Precipitation	x	-	-
Evapotranspiration	x	x	x

1) Considered only in the National application (multi layer models)
2) Considered in the European - but not in the National application
3) Considered in the National - but not in the European application

Location and soil type will certainly influence litterfall - and root decay fluxes but information has been considered too scarce to include these effects.

In order to get data for all forest ecosystems within all grids, a first good approach is to interprete and extrapolate available data by deriving relationships (transfer functions) between the data mentioned before and basic land and climate characteristics, such as tree species, soil type, elevation, precipitation, temperature etc., which are available in geographic information systems.

A summarizing overview of the data acquisition strategy for the inputs to and outputs from the soil system is given in Table 9.

Table 9 Data acquisition strategies for inputs and outputs to the soil system

Inputs	Outputs	Elements	Data aquisition strategy
Total deposition	-	SO_2, NO_x, NH_3	**Estimate per grid using emission/deposition matrices, corrected for forest filtering**
Wet deposition	-	**Ca, Mg, K, Na**	**Estimate per grid based on data of weatherstations or monitoring sites**
Dry deposition	-	**Ca, Mg, K, Na**	**Derivation of a transfer function with wet deposition and forest coverage**
-	**Foliar uptake**	NH_4, NO_3, SO_4	**Derivation of a transfer function with total deposition and tree species**
Foliar exudation	-	**Ca, Mg, K**	**Estimate for each tree species based on literature data**
Litterfall	-	**N, Ca, Mg, K, S**	**Estimate for each tree species based on literature data**
Root turnover	-	**N, Ca, Mg, K, S**	**Estimate for each tree species based on literature data**
-	**Net uptake**	**N, Ca, Mg, K, S**	**Derivation of a transfer function with location, tree species and soil type**
Precipitation	-	-	**Estimate per grid based on data of weatherstations or monitoring sites**
-	**Interception**	-	**Derivation of a transfer function with precipitation amount and tree species**
-	**Transpiration**	-	**Calculation as a function of climate, tree species and soil type**

A more detailed overview of the various methods to derive data on inputs to - and outputs from the soil system on a European and National scale is given in the following subparagraphs.

5.1.2 Atmospheric deposition

Sulphur and nitrogen deposition in Europe

For the European application, total deposition estimates for SO_2, NO_x and NH_3 are derived from emission deposition matrices within the RAINS model (Alcamo et al., 1987) for a grid square of 1.0° longitude x 0.5° latitude. These matrices are based on EMEP model calculations. The EMEP-model (Eliassen et al., 1988) calculates the total (both wet and dry) deposition of sulphur of nitrogen on a European scale for a grid of 150 x 150 km^2. Estimates are calibrated on available monitoring data about wet deposition and air pollutant concentrations from the EMEP network. Dry deposition is not determined on a routine basis since the eddy correlation and gradient methods that are generally used in this respect require sophisticated equipment and extentive monitoring efforts.

The estimates of either the EMEP model or emission-deposition matrices do give information about the total deposition of SO_2, NO_x and NH_3 on a grid. However, there is a large variation within each grid due to a large difference in dry deposition on various receptors. For example, dry deposition on a conifereous stand is generally twice as large as on a heathland vegetation. There are also differences in filtering dry deposition between coniferous and deciduous stands and even between tree species.

The influence of forest filtering on the deposition is included by relating the deposition on coniferous and deciduous forests, dt_c and dt_d, to the average deposition on a grid, dt_g, according to:

$$dt_c = ff_c \cdot dt_g \qquad (128)$$

$$dt_d = ff_d \cdot dt_g \qquad (129)$$

where ff_c and ff_d are filtering factors (-) for both coniferous and deciduous forests, respectively.

When dt_c and dt_d are calculated higher than dt_g, then the deposition on non-forested land must be smaller if the average total deposition on the grid is assumed correct. Theoretically, this might give rise to unrealistically low depositions on open land, e.g. below the wet deposition or even below zero. In order to avoid this, it is assumed that the total deposition on open land, dt_o, is at least as high as the wet deposition, dw_g.

Calculating the deposition on open land as:

$$dt_o = ff_o \cdot dt_g \qquad (130)$$

where ff_o is the filtering factor for open land, this implies that:

$$ff_o \geq \frac{dw_g}{dt_g} \qquad (131)$$

Assuming that the total deposition on a grid square is correct which means that:

$$dt_g = f_c \cdot dt_c + f_d \cdot dt_d + f_o \cdot dt_o \qquad (132)$$

and combining the Equations (128), (129), (130) and (132) gives:

$$ff_o = (1 - f_c \cdot ff_c - f_d \cdot ff_d) / f_o \tag{133}$$

where f_c, f_d and f_o are the coverage fractions of coniferous forests, deciduous forests and open land (-) respectively, with:

$$f_o = 1 - f_c - f_d \tag{134}$$

When a value for ff_o less than that given by the ratio of wet to total deposition is obtained, then ff_o is set to the value given by this ratio and ff_c and ff_d are recalculated. This is done by inserting the new value of ff_o into Equation (128) and assuming that there is a constant ratio between the forest filtering factors ff_c and ff_d, R_{cd}, i.e.

$$ff_c = R_{cd} \cdot ff_d \tag{135}$$

Combining the Equations (130), (133) and (135) yields

$$ff_d = \frac{1 - f_o \cdot (dw_g / dt_g)}{f_c \cdot R_{cd} + f_d} \tag{136}$$

For given values of the ratio of wet to total deposition per grid, both ff_d and ff_c can be calculated with the Equations (135) and (136) since the forest coverage fractions (f_c, f_d and f_o) per grid are known.

Values for ff_d and ff_c (and R_{cd}) on a European scale can be derived from Ivens et al. (1989) who compared throughfall data for SO_4, NO_3 and NH_4 with total deposition estimates using the EMEP model. Values of ff_d thus derived equal 1.0 for SO_2, 0.7 for NO_x and 1.2 for NH_3 whereas R_{cd} equals 1.6 for all elements.

Sulphur and nitrogen deposition in the Netherlands

Sulphur and nitrogen deposition on a national scale can be derived from TREND model calculations that give information about the average deposition on a grid. (Van Jaarsveld, 1989). For national applications, total deposition estimates for SO_2, NO_x and NH_3 are derived by using emission-deposition matrices within the DAS model (Olsthoorn et al., 1990) that are based on TREND model calculations.

As with the European application, the influence of forest filtering is included by multiplying the deposition on the grid by filtering factors for spruce forests (Douglas Fir and Norway Spruce), pine forests (Scotch Pine and Black Pine) and deciduous forests (including Japanese Larch) according to:

$$dt_s = ff_s \cdot dt_g \tag{137}$$

$$dt_p = ff_p \cdot dt_g \tag{138}$$

$$dt_d = ff_d \cdot dt_g \tag{139}$$

where ff_s, ff_p and ff_d are the filtering factors (-) for spruce forests, pine forests and deciduous forests respectively. Analogous to Equation (136) a new value of ff_d is calculated when ff_o is less than the ratio of wet to total deposition, according to:

$$ff_d = \frac{1 - f_o \cdot dw_g/dt_g}{f_s \cdot R_{sd} + f_p \cdot R_{pd} + f_d} \tag{140}$$

where f_s and f_p are the coverage fractions (-) of spruce forests and pine forests respectively and R_{sd} and R_{pd} are the ratios between the forest filtering factors (-) for spruce - and deciduous forest and pine - and deciduous forest respectively. However, in the Netherlands, recalculation of the filtering factors is hardly ever necessary since forests generally cover only a small fraction of the land area in a grid.

Values for ff_s, ff_p and ff_d can be derived from a comparison of throughfall data below spruce -, pine - and deciduous forests in the Netherlands (42 sites; Duysings et al., 1986; Van Breemen et al., 1986; Ivens et al., 1988; Kleijn et al., 1989; Tiktak et al., 1988; Houdijk, 1990) and total deposition estimates with the TREND model. Values thus derived are given in Table 10.

Table 10 Forest filtering factors for SO_4, NH_4 and NO_3 for spruce -, pine - and deciduous forests in the Netherlands.

Forest type	**Filtering factors (-)**		
	SO_4	**NH_4**	**NO_3**
Spruce forest	**1.60**	**1.50**	**1.00**
Pine forest	**1.40**	**1.30**	**0.85**
Deciduous forest	**1.15**	**1.10**	**0.70**

The values of R_{sd} and R_{pd} are 1.4 and 1.2 respectively for all components (cf Table 10).

Base cation deposition

The total deposition of deposition base cations and Cl can partly be derived from weather stations, which give bulk deposition data. Bulk deposition data for each receptor area (grid) are derived from 22 weatherstations in the Netherlands (KNMI, 1985) and 76 stations in Europe (Schaug et al., 1986), using interpolation techniques to get values for each grid according to:

$$BC_{dw,i} = \sum_{j=1}^{n} \left(\frac{1}{r_{i,j}}\right) \cdot BC_{dw,j} \Big/ \sum_{j=1}^{n} \left(\frac{1}{r_{i,j}}\right) \tag{141}$$

where $BC_{dw,i}$ and $BC_{dw,j}$ are the bulk (wet) depositon of base cations in grid i and at weatherstation j respectively, $r_{i,j}$ is the distance of the centre of grid i to weatherstation j and n is the number of weatherstations.

However, bulk deposition data mainly includes wet deposition (a very small part is dry deposition). The influence of dry deposition on the total deposition can be accounted for by multiplying the bulk (wet) deposition according to:

$$BC_{dt} = (1 + f_{dd}) \cdot BC_{dw} \tag{142}$$

where f_{dd} is a dry deposition factor (-).

Equation (142) is based on the implicit assumption that dry deposition is linearly related to wet deposition. The value of f_{dd} is derived from the ratio of Na in bulk deposition and throughfall according to Ulrich, 1983; Bredemeier, 1988):

$$f_{dd} = (Na_{tf} - Na_{dw}) / Na_{dw} \tag{143}$$

Results of a literature survey by Ivens et al (1989) for 47 sites in Europe give median values for f_{dd} of 0.6 for deciduous forests and 1.1 for coniferous forests. However, these data are based on results in areas which are sparsely occupied by forests. It is to be expected that the dry deposition factor f_{dd} will decrease with an increase in the forested areas within a grid. For the application on Europe, this effect is accounted for by a linear relationship between f_{dd} and the fraction of open land, f_o, according to:

$$f_{dd} = \alpha \cdot f_o \tag{144}$$

where α = 0.6 for deciduous forests and 1.1 for coniferous forests.

In the Netherlands, f_o is nearly always close to 1 and f_{dd} is considered independent of the forest coverage. As with forest-filtering, values are derived from a comparison of Na in throughfall and bulk deposition at 42 sites in the Netherlands. Values thus derived equal about 2.0 for spruce forests, 1.5 for pine forests and 1.0 for deciduous forests.

5.1.3 Foliar uptake and exudation

Foliar uptake and - exudation is only included in the models MACAL and RESAM, which are developed for application in the Netherlands. Foliar uptake fractions for H and NH_4 are assumed to be equal (cf section 3.4.3) and are calculated on the basis of the total deposition of H and NH_4 and the foliar excudation of Ca, Mg and K according to:

$$frH_{fu} = frNH_{4,fu} = \frac{Ca_{fe} + Mg_{fe} + K_{fe}}{H_{dt} + NH_{4,dt}} \tag{145}$$

The foliar exudation of Ca, Mg and K is calculated for the 42 Dutch forest sites mentioned before from the difference between throughfall and total deposition calculated according to Equation (142) and (143). The total deposition of NH_4 and H is based on the throughfall data of these ions on these sites. This is most likely an underestimate due to foliar uptake. Foliar uptake fractions for SO_4 and NO_3 are also derived from bulk and throughfall data using a procedure described in Van der Maas et al. (in prep.).

Foliar exudation fractions for Ca, Mg and K are simply derived from a comparison of Ca, Mg or K exudation to the total exudation of these cations (cf Equation (75)). Values thus derived are given in Table 11.

***Table 11** Foliar uptake and foliar exudation fractions for coniferous and deciduous forests in the Netherlands*

Forest type	Foliar uptake fraction (-)			Foliar exudation fraction (-)		
	NH_4	NO_3	SO_4	Ca	Mg	K
Coniferous forest	0.30	0.05	0.10	0.24	0.13	0.63
Deciduous forest	0.30	0.05	0.10	0.18	0.16	0.66

5.1.4 Litterfall and root turnover

In the multi-layer model RESAM, nutrient cycling by litterfall and root-turnover plays an important role. In this model the element flux due to these processes is calculated by multiplication of the turn-over constants, amounts and element contents in leaves (needles) and fine roots (cf Equation (76) and (106)). Data for these parameters, for major tree species in the Netherlands, are given in Table 12.

Data on the average biomass and turnover constants and the element contents in roots are based on a literature compilation by De Vries et al. (1990). Data on the element contents in leaves are based on an inventory of 150 forest stands in the Netherlands in 1990 (Van den Burg, pers. comm.).

***Table 12** Average values for the biomass, turnover constants and the Ca, Mg and K content in leaves (needles) and fine roots of five coniferous and two deciduous tree species*

Tree species	Biomass (kg ha^{-1})		Turnover constants (yr^{-1})		Content in leaves (%)			Content in fine roots (%)		
	Leaves	Fine roots	Leaves	Fine roots	Ca	Mg	K	Ca	Mg	K
Scotch Pine	5500	5000	0.55	1.4	0.24	0.07	0.60	0.13	0.05	0.15
Black Pine	7250	5000	0.35	1.4	0.12	0.08	0.59	0.06	0.04	0.13
Douglas Fir	10850	4700	0.28	1.4	0.40	0.14	0.61	0.21	0.02	0.05
Norway Spruce	16600	5650	0.20	1.4	0.27	0.08	0.54	0.27	0.07	0.34
Japanese Larch	4350	5200	1.0	1.4	0.42	0.18	0.79	0.27	0.07	0.34
Oak	3300	5500	1.0	1.4	0.49	0.15	0.92	0.27	0.09	0.35
Beech	2850	6500	1.0	1.4	0.52	0.11	0.72	0.12	0.03	0.16

The MACAL model only includes litterfall. This model will also be applied for five additional deciduous trees, i.e. Poplar, Willow, Birch, Ash and Black Alder. Data for these tree species are given in Table 13.

***Table 13** Indicative values for the biomass and Ca, Mg and K contents in the leaves of five deciduous tree species*

Tree species	Biomass ($kg\ ha^{-1}$)	Element contents (%)		
		Ca	Mg	K
Poplar[1)]	4000	1.17	0.21	1.07
Willow[1)]	3500	1.07	0.21	0.85
Birch[2)]	2500	0.98	0.25	0.93
Ash[1)]	2500	0.81	0.28	0.97
Black Alder[2)]	4000	0.98	0.23	1.15

1) Derived from Kimmins et al. (1985)
2) Derived from a recent inventory in the Netherlands (Eelerwoude, 1991)

In order to get values in $mol_c\ kg^{-1}$, the data on elements contents in Table 12 and 13 have to be divided by 2.0 for Ca, 1.2 for Mg and 3.9 for K.

Both in MACAL and RESAM, the nitrogen content in leaves and roots is calculated as a function of the N deposition according to:

$$ctN = ctN_{min} + \alpha \cdot (ctN_{max} - ctN_{min}) \tag{146}$$

where ctN_{min} and ctN_{max} are the minimum and maximum N content in leaves and fine roots ($mol_c\ kg^{-1}$).

The value of α is calculated according to:

$$\alpha = \frac{N_{dt} - N_{dt,min}}{N_{dt,max} - N_{dt,min}} \qquad \text{for } N_{dt,min} < N_{dt} < N_{dt,max} \tag{147}$$

$$\alpha = 0 \qquad \text{for } N_{dt} < N_{dt,min} \tag{148}$$

$$\alpha = 1 \qquad \text{for } N_{dt} > N_{dt,max} \tag{149}$$

where $N_{dt,min}$ and $N_{dt,max}$ are the minimum and maximum values between which the N deposition influences the N content of leaves. Values used for Ndt_{min} and Ndt_{max} are 1500 and 7000 mol $ha^{-1}\ yr^{-1}$ respectively, based on data given by Van den Burg et al. (1988) and Van den Burg and Kiewiet (1989).

Values for the minimum and maximum N content in leaves and roots, (to be) used in the regional application of MACAL and RESAM are given in Table 14. Data are based on literature compilations of Rodin and Bazilevich (1967), Kimmins et al. (1985) and De Vries et al. (1990).

Table 14 Values for the minimum and maximum N content in leaves (needles) and fine roots of coniferous and deciduous tree species

Tree species	N content in leaves (%)		N content in fine roots (%)	
	min.[1)]	max.	min.[1)]	max.
Scotch Pine	1.0 (2.0)	3.5	0.4 (0.7)	1.0
Black Pine	1.0 (2.0)	2.5	0.4 (0.7)	1.0
Douglas Fir	1.0 (2.0)	3.5	0.4 (0.7)	1.0
Norway Spruce	1.5 (2.5)	3.5	0.5 (1.0)	1.5
Japanese Larch	1.5 (2.5)	3.5	0.5 (1.0)	1.5
Deciduous trees[2)]	1.5 (2.5)	3.5	0.5 (1.0)	1.5

1) Values between brackets are the minimum contents above which reallocation occurs (cf Equation (78)). Maximum contents are equal.

2) Oak, Beech, Poplar, Willow, Birch, Ash and Black Alder

5.1.5 Net uptake

Calculation

The amount of nitrogen and base cations removed in harvesting depends upon tree species, forestry practice and site quality. With respect to harvesting both stemwood removal and whole-tree harvesting (stem and branches) is practiced.

The average annual element uptake in stems and branches, can be derived by multiplying the annual increase in biomass with the element contents in the various compartments according to:

$$X_{gu} = k_{gr} \cdot \rho_{st} \cdot (ctX_{st} + f_{br,st} \cdot ctX_{br}) \tag{150}$$

where X_{gu} is the net uptake flux of element X ($mol_c\ ha^{-1}\ yr^{-1}$), k_{gr} is the annual average growth rate constant ($m^3\ ha^{-1}\ yr^{-1}$), ρ_{st} is the density of stemwood ($kg\ m^{-3}$), (which is assumed equal to the density of branch wood), ctX_{st} is the content of element X in stems ($mol_c\ kg^{-1}$), ctX_{br} is the content of element X in branches ($mol_c\ kg^{-1}$) and $f_{br,st}$ is the branch to stem ratio ($kg\ kg^{-1}$). The contribution of branches can be neglected in case of stemwood removal only.

Europe

On a European scale there is only a differentiation between coniferous and deciduous trees (section 4.2.1). Data on average stem growth rates in Europe can be derived from a literature compilation by Nilson and Sallnäs (in prep.). These data will be used to get average growth rate estimates for coniferous and deciduous forests in each 1.0° longitude x 0.5° latitude grid over Europe, excluding the boreal forests area. In this area the growth rate is calculated from a relationship with the temperature according to (Kauppi and Posch, 1985):

$$k_{gr} = k_{gr,max} / (1 + e^{-\alpha ETS + \beta}) \tag{151}$$

where $k_{gr,max}$ is the maximum growth rate constant ($m^3\ ha^{-1}\ yr^{-1}$) and ETS is the effective temperature sum, i.e. the annual summation of temperature for all days with a temperature above 5°C (°C). Values that are used for $k_{gr,max}$, α and β are 6.0, 0.005 and 5 respectively (Kauppi and Posch, 1985).

Data for the density of stemwood and the branch to stem ratio can be derived from the data for the various tree species. Average values used are 500 kg m^{-3} and 700 kg m^{-3} for the stemwood density and 0.15 kg kg^{-1} and 0.20 kg kg^{-1} for the branche to stem ratio of coniferous and deciduous forests respectively Kimmins et al., 1985; De Vries et al., 1990).

Data for the element contents in stems and branches are related to the latitude according to:

$$ctX = ctX_{min} + \alpha\ (ctX_{max} - ctX_{min}) \tag{152}$$

where ctX_{min} and ctX_{max} are the minimum and maximum contents ($mol_c\ kg^{-1}$) of element X in stems or branches.

For X = N, Mg and K, α is equal to:

$$\alpha = \frac{65 - \text{latitude}}{65 - 55} \qquad \text{for } 55° < \text{latitude} < 65° \tag{153}$$

$$\alpha = 0 \qquad \text{for latitude} \geq 65° \tag{154}$$

$$\alpha = 1 \qquad \text{for latitude} \leq 55° \tag{155}$$

For X is Ca, α is equal to:

$$\alpha = \frac{\text{latitude} - 55}{65 - 55} \qquad \text{for } 55° < \text{latitude} < 65° \tag{156}$$

$$\alpha = 0 \qquad \text{for latitude} \leq 55° \tag{157}$$

$$\alpha = 1 \qquad \text{for latitude} \geq 65° \tag{158}$$

Using these Equations, element contents in stems and branches of boreal forests (above latitude 55°) are either lower (N, Mg and K) or higher (Ca) than in Central and Southern European forests (below latitute 55°). The relation with latitude in boreal forests is based on available data given in Rosén (1990). Values for the minimum and maximum element contents are given in Table 15.

Table 15 Minimum and maximum values of nitrogen and base cation contents in stems and branches of coniferous and deciduous forests in Europe

Forest type	Compartment	Minimum contents (%)				Maximum contents (%)			
		N	Ca	Mg	K	N	Ca	Mg	K
Coniferous	Stems	0.10	0.08	0.02	0.05	0.10	0.16	0.02	0.05
	Branches	0.20	0.30	0.03	0.10	0.40	0.60	0.05	0.25
Deciduous	Stems	0.15	0.13	0.04	0.10	0.15	0.21	0.04	0.10
	Branches	0.20	0.45	0.03	0.05	0.40	0.75	0.05	0.20

Except for calcium, latitude does not affect the element contents in stemwood (cf Table 15). The minimum values for Ca and the maximum values of N, Mg and K apply to all forests below latitude 55°. Constant values below this latitude does not imply that there is no influence of latitude, but that there are no readily available data to derive a relationship.

The Netherlands

For the Netherlands, more detailed information exists on the average growth rate. Here, values have been derived as a function of both tree species and soil type (De Vries et al., 1990). For each combination of tree species and soil type a suitability class (good, medium or low) was defined (Hendriks, pers. comm.) for which an average growth rate has been assigned (Table 16).

Table 16 Average growth rates of twelve tree species in the Netherlands for three suitability classes

Tree species	Growth rates ($m^3\ ha^{-1}\ yr^{-1}$)			
	good	medium	low	average
Scotch Pine[1)]	7.1	5.5	3.1	6.3
Black Pine[1)]	10.6	7.6	5.0	9.2
Douglas Fir[1)]	14.7	11.1	6.6	10.9
Norway Spruce[1)]	13.6	8.9	5.0	7.9
Japanese Larch[1)]	14.0	10.9	5.7	10.3
Oak[1)]	8.0	6.0	4.0	6.3
Beech[2)]	7.0	5.0	3.0	5.1
Poplar[3)]	19.7	14.0	10.8	17.4
Willow[3)]	14.8	11.9	6.7	14.2
Birch[2)]	8.0	6.0	4.0	6.0
Ash[2)]	8.0	5.0	3.0	7.0
Black Alder[4)]	8.5	5.0	4.5	7.7

1) Derived from La Bastide and Faber (1972)
2) Derived from Hamilton and Cristie (1971)
3) Derived from Faber and Thiemens (1975)
4) Derived from Van den Burg (1978)

The average values in Table 16 have been calculated from the occurence of the various tree species on the different suitability classes (soil type dependent). A recent investigation on the (average) growth rates of Dutch forests give nearly similar values except for Beech for wich the average actual value appears to be much higher, i.e. 12.8 $m^3\ ha^{-1}\ yr^{-1}$ (Houtoogst, 1991).

Average data on the density of stemwood, the branche to stem ratio and the element contents in stems and branches of major Dutch tree species have been compiled in De Vries et al (1990). Average data thus derived are given in Table 17.

Table 17 Average values for the density of stemwood, the branche to stem ratio and the base cation content in stems and branches of coniferous and deciduous tree species

Tree species	Density ($kg\ m^{-3}$)	Ratio ($kg\ kg^{-1}$)	Stem contents (%)			Branche contents (%)		
			Ca	Mg	K	Ca	Mg	K
Scotch Pine	**510**	**0.15**	**0.09**	**0.02**	**0.05**	**0.19**	**0.04**	**0.21**
Black Pine	**510**	**0.30**	**0.06**	**0.02**	**0.05**	**0.42**	**0.07**	**0.22**
Douglas Fir	**530**	**0.10**	**0.07**	**0.01**	**0.04**	**0.50**	**0.06**	**0.26**
Norway Spruce	**460**	**0.15**	**0.14**	**0.02**	**0.07**	**0.23**	**0.08**	**0.37**
Japanese Larch	**550**	**0.15**	**0.06**	**0.01**	**0.04**	**0.23**	**0.08**	**0.37**
Oak	**700**	**0.30**	**0.10**	**0.02**	**0.13**	**0.42**	**0.03**	**0.18**
Beech	**700**	**0.25**	**0.11**	**0.03**	**0.10**	**0.24**	**0.02**	**0.13**
Poplar	**450**	**0.20**	**0.15**	**0.02**	**0.08**	**0.60**	**0.08**	**0.25**
Willow	**450**	**0.15**	**0.09**	**0.03**	**0.07**	**0.59**	**0.07**	**0.32**
Birch	**650**	**0.15**	**0.17**	**0.03**	**0.08**	**0.54**	**0.05**	**0.15**
Ash	**580**	**0.15**	**0.08**	**0.03**	**0.13**	**0.50**	**0.07**	**0.24**
Black Alder	**530**	**0.15**	**0.09**	**0.03**	**0.07**	**0.59**	**0.07**	**0.32**

As with the leaves (needles) and fine roots, the N content in stems and branches is calculated in the MACAL and RESAM model according to the Equations (146) to (149). Values for the minimum and maximum N content in stems and branches, (to be) used in the regional application of MACAL and RESAM are given in Table 18. Data are based on the literature compilations by Kimmins et al. (1985) and De Vries et al. (1990).

Table 18 Values for the minimum and maximum N content in stems and branches of coniferous and deciduous tree species

Tree species	N content in stems (%)		N content in branches (%)	
	min.	max.	min.	max.
Scotch Pine	**0.08**	**0.20**	**0.20**	**0.50**
Black Pine	**0.05**	**0.15**	**0.20**	**0.50**
Douglas Fir	**0.08**	**0.20**	**0.20**	**0.50**
Norway Spruce	**0.08**	**0.20**	**0.35**	**0.75**
Japanese Larch	**0.08**	**0.20**	**0.35**	**0.75**
Deciduous trees[1)]	**0.15**	**0.25**	**0.35**	**0.75**

1) Oak, Beech, Poplar, Willow, Birch, Ash and Black Alder

5.1.6 Precipitation and evapotranspiration

Europe

Similar to the base cation deposition, data for the precipitation on each grid can be derived from weather stations in Europe. A thorough review of such data, available in several global databases, is given in Leemans and Cramer (1990). Selected records of average monthly precipitation from 1678 stations, during the period 1930-1960, have been interpolated by these authors, to the 1.0° longitude x 0.5° latitude grid. Details on the selection and interpolation procedure are given in Leemans and Cramer (1990). Yearly averaged values for this period are used in the European aplication of START and SMART. Precipitation ranges mostly between 400 and 700 mm yr^{-1} in the Northern and Eastern part of Europe (Sweden, Finland, Poland, Czechoslovakia, Hungary, Romania, Bulgaria and the USSR) and in Spain and between 700 and 1500 mm yr^{-1} in Central and Southern Europe including large parts of the UK and Norway. In the latter countries and in the Alps values can become as large as 2850 mm yr^{-1}.

For the European application with START and SMART an average actual evapotranspiration for forests in a grid (ET_f) is derived from a database at IIASA. Data of ET_f have been calculated with a simple model, as the daily integral of the lesser of a supply and demand function (Cramer and Prentice, 1988). The supply is linearly related to soil moisture, whereas the demand is calculated as a function of temperature, cloudiness and latitude with the Penman-Monteith model. Details of the application procedure are given in Cramer and Prentice (1988). Evapotranspiration data thus derived vary mostly between 300 and 500 mm yr^{-1} in the Scandinavian countries and the USSR, including large parts of the UK and Spain and between 500 and 700 mm yr^{-1} in Central and Southern Europe.

The influence of forest type on the evapotranspiration value is accounted for by a procedure similar to forest filtering. Actual evapotranspiration ET_a is a lumped expression for the sum of interception evaporation I, actual soil evaporation E_a and actual transpiration T_a (cf Equation (22)). The values of I and E determine the infiltration at the soil surface. The average actual evapotranspiration for forests in a grid (ET_f), is assumed correct, whereas differences between coniferous and deciduous forests are assumed to be caused by interception evaporation only. In other words, the sum of evaporation, E and transpiration, T, denoted as ET_f is assumed indepent of tree species which is reasonable (Roberts, 1983). On a grid basis, this value can be calculated as

$$ET'_f = ET_f - I_f \tag{159}$$

where I_f is the average interception evaporation (mm yr^{-1}) for forests in a grid, which is calculated as

$$I_f = \left(\frac{f_c}{f_c + f_d}\right) \cdot I_c + \left(\frac{f_d}{f_c + f_d}\right) \cdot I_d \tag{160}$$

where I_c and I_d are the interception evaporation values for coniferous forests and deciduous forests (mm yr^{-1}) respectively. The evapotranspiration for each receptor can be calculated by adding I_c and I_d, respectively, to ET'_f.

Interception of rainwater by the forest canopy depends upon the forest type (tree species) and the precipitation amount. On the basis of data given by Leyton et al. (1967) and Calder and Newson (1979), interception evaporation can be described as a function of precipitation (P) according to:

$$I = a \cdot P^{0.75} \tag{161}$$

The value of a depends upon land use and equals about 1.75 for coniferous forests (Mitscherlich and Moll, 1970) and 1.0 for deciduous forests (Van Grinsven, 1987). The maximum yearly interception evaporation thus equals 680 and 390 mm yr^{-1} for coniferous - and deciduous forests respectively using a maximum precipitation rate of 2850 mm yr^{-1}. Interception evaporation data for high rainfall areas in Scotland are consistent with these values (Cape and Lightowlers, 1988).

The precipitation surpluses thus derived varies mostly between 20 and 200 mm yr^{-1} in the Northern and Eastern European countries with a precipitation rate below 700 mm yr^{-1} (see before). In Central and Southern Europe most values range between 100 and 300 mm yr^{-1}. Extremely high values above 1000 mm yr^{-1} occur mainly in Norway, Scotland, Ireland, Switzerland and Austria.

The Netherlands

Precipitation data can be derived from weather stations from the Royal Netherlands Meteorological Institute (KNMI). Selected records of precipitation normals from 280 stations over the period 1950-1980 have been interpolated to each 10 x 10 km grid. Details on the interpolation procedure are given in Hootsmans and Van Uffelen (1991). Most values range between 700 and 900 mm yr^{-1}.

Interception evaporation is calculated as a fraction of the precipitation. This fraction decreases with an increasing precipitation amount (cf Equation (161)), but the range in precipitation in the Netherlands is considered to small to include this effect. Average values used for the interception evaporation fraction are 0.45 for Norway Spruce; 0.40 for Douglas Fir; 0.30 for Scotch Pine, Black Pine and Beech; 0.25 for Japanese Larch, Poplar and Ash and 0.20 for Oak, Willow, Birch and Black Alder. Data are based on a literature compilation by Hiege (1985) except for Black Pine, Ash, Willow and Black Alder. Values for these tree species are based on data from tree species with comparable physical tree characteristics such as canopy storage capacity (Hendriks, pers. comm.). Interception evaporation values thus derived vary mostly between 150 mm yr^{-1} for deciduous forests up to 350 mm yr^{-1} for Norway Spruce.

Soil evaporation data are based on calculations with the simulation model SWATRE (Belmans et al., 1983). The average value equals 50 mm yr^{-1} (De Visser and De Vries, 1989).

Transpiration data are based on model calculations with SWATRE, using precipitation data of a mean hydrological year (De Visser and De Vries, 1989). Calculations have been limited to Scotch Pine, Douglas Fir and Oak on non-calcareous sandy soils (cf Table 7). Transpiration values for Black Pine are assumed equal to those of Scotch Pine, whereas values for Japanese Larch, Beech, Poplar, Willow, Birch, Ash and Blak Alder are assumed equal to those of Oak. Norway Spruce is those assumed to transpire 10% more than Douglas Fir (Hendriks, pers. comm.). Data for loess soils are assumed equal to those of fine textured leptic podzols. For clay and peat soils potential transpiration values are taken. Values thus derived nearly all range between 250 and 350 mm yr^{-1} except for wet soils. This is in accordance with data from Roberts (1983) who found that the actual transpiration of forests in most European countries equals approximately 300 mm yr^{-1}, independent of tree species and soil type. An overview of the transpiration data is given in Hootsmans and Van Uffelen (1991). The precipitation surplus thus derived ranges mostly between 50 and 250 mm yr^{-1} for coniferous forests and between 150 and 300 mm yr^{-1} for deciduous forests.

The MACAL and RESAM model also require data on the distribution of transpiration with depth. This is determined by the distribution of fine roots (< 2 mm), which are mainly responsible for the uptake of water and nutrients. Literature data show that these roots mainly occur in the topsoil i.e. the litter layer and the top 20 cm of the mineral soil (Grier et al., 1981; McClaugherty et al., 1982; Persson, 1983; Harris et al., 1977; Kimmins and Hawkes, 1978). Generally 20-50% of the fine roots occur in the litter layer. A reasonable overall estimate for the distribution of fine roots in pine forests, based on the literature given above is: litterlayer (35%), 0-10 cm (30%), 10-20 cm (20%), 20-30 cm (10%), > 30 cm (5%). In deciduous forests, the distribution migth be more even. When the soil is reworked, which is often the case in the Netherlands, fine roots are more evenly distributed (Oterdoom et al., 1991). The distribution of fine roots, and the resulting distribution of transpiration, for the various combinations of tree species and soil types included in the RESAM application, are given in De Visser and De Vries (1989). In MACAL the root uptake pattern is set linear, i.e. a root uptake coefficient (n in Equation (90)) of 2.0 is used.

5.2 Soil data

5.2.1 Data acquisition strategy

The soil data that are needed when using a dynamic soil acidification model are given in Table 19. Data are divided in soil variables, soil properties and soil constants.

Table 19 Soil data needed in the models SMART and RESAM

Type of soil data	Data
Soil variables	
Element contents in the litter layer	Contents of C, N, Ca, Mg, K and S in litter
Element contents in minerals	Carbonate contents
	Total contents of Ca, Mg, K and Na[1)]
	Al-hydroxide contents
Element contents at the adsorption complex	Fractions of H, Al, Ca, Mg, K[1)], Na[1)] and NH_4[1)] on the adsorption complex
Element contents in the soil solution	Concentrations of H, Al, Ca, Mg, K[1)], Na[1)], NH_4, NO_3, SO_4, Cl[1)], HCO_3 and RCOO[1)]
Soil properties	Bulk density and soil thickness
	Cation exchange capacity
	Sulphate sorption capacity
Soil constants	
Equilibrium constants	Exchange constants for H, Al, Mg, NH_4[1)], K[1)] and Na[1)] against Ca
	Adsorption constant for SO_4
	Dissociation constant for CO_2
	Equilibrium constants for the dissolution of $CaCO_3$ and $Al(OH)_3$[2)]
Rate constants	Mineralization rate fractions and constants
	Nitrification and denitrification rate fractions[2,3)]
	Weathering rate constants[2)]
	Rate constants for the dissolution of $CaCO_3$ and $Al(OH)_3$[1)]

1) These data are only needed in RESAM and not in SMART
2) These data are also needed in the steady-state models START and MACAL
3) RESAM needs rate constants

As with the inputs and outputs to the soil system, soil data for all (major) forest soil types in all grids are mainly derived by extrapolation of point data by using transfer functions between model inputs such as CEC, base saturation etc. and basic land and soil characteristics such as soil type, soil horizon, organic matter contents, soil texture, etc. (De Vries, 1987; De Vries et al., 1989c). Exceptions are several soil variables and equilibrium constants that are derived from available literature.

An overview of the various transfer functions and data on soil variables, soil properties and soil constants is given in the folowing subparagraphs. When new data are to be obtained, various sampling strategies can be used. This is illustrated in an appendix.

5.2.2 Soil variables

Soil variables include element contents in litter, minerals, at the adsorption complex and in the soil solution (Table 19).

Europe

The SMART model only requires data on the N content in litter, the carbonate and Al-hydroxide content, the fraction of BC, Al and H at the adsorption complex and the concentration of BC, Al and H in solution (Table 19).

The N content in litter is related to the N deposition according to the Equations (146) to (149). Values for the minimum and maximum N content in litter are taken equal to 1.25 and 2.5%. This is based on N contents in litter in Dutch forests in 1940 (De Vries and Van Vliet, 1945) and at present (Kleijn et al., 1989). The lower value can also be derived from litter data in relatively unpolluted areas such as Finland (Mälkönen et al., 1991). Using a C content of 50%, this is equivalent to a C/N ratio of 40 and 20 respectively. This is also the range between which N immobilization is assumed to occur, i.e. C/N_{cr} is taken at 40 and C/N_m at 20. The minimum and maximum value for N deposition are taken equal to 350 and 3500 $mol_c\ ha^{-1}\ yr^{-1}$, representing the average deposition levels in unpolluted and highly polluted areas respectively.

Data for the carbonate and Al-hydroxide content are based on information in the FAO soil map of Europe (FAO, 1981). Data only refer to one selected profile for each soil type. It is aimed to develop a much more detailed soil information system for Europe on the basis of available data in the various countries.

The initial base saturation of soils is calculated as the maximum of either (1) a relation with the texture class of soils (Table 20) or (2) an equilibrium with present deposition levels of SO_4, NO_3, NH_4 and BC. Information on the various texture classes is given in section 4.2.1.

Table 20 Base saturation (%) in European forest soils as a function of texture class

Class 1	Class 1/2	Class 1/3	Class 2	Class 2/3	Class 3
5	10	10	15	40	50

The relation (class transfer function) in Table 20 is based on data from forest soils given in FAO (1981) and in Gardiner (1987). An increase in clay content implies an increase in weathering rate, which implies an increase in base saturation. For histosols (peat soils) the initial base saturation is put equal to 70% for a Eutric Histosol (Oe) and 10% for a Dystric Histosol (Od). The Al and H saturation is calculated by the SMART model as given in De Vries et al. (1989a).

The base saturation and Al and H saturation in equilibrium with present deposition levels are derived by calculating the soil solution composition according to the START model (by combining the charge balance Equation (37) with the mass balance Equations for SO_4, NO_3, NH_4 and BC (38) and the equilibrium Equations for Al (40) and HCO_3 (41)) and inserting the values for [H], [Al] and [BC] in the various exchange Equations (100) to (102). Especially in southern European countries, where the acid deposition is relatively low and the base cation input is high, the base saturation in equilibrium with the present load can be higher than the value assigned according to Table 20.

The initial concentrations of SO_4, NO_3 and NH_4 are always calculated from the present annual atmospheric input and the precipitation surplus. When the base saturation is in equilibrium with the present deposition levels, concentrations of H, Al, BC and HCO_3 are derived by combining the charge balance Equation (37), the mass balance Equation for BC (38) and the equilibrium Equations for Al (40) and HCO_3 (41). Otherwise, these concentrations are derived by combining the same set of Equations with the exchange Equations for H, Al and BC ((100) to (102)).

The Netherlands

The RESAM model requires data on all the soil variables mentioned in Table 19.

Initial element contents in litter are taken equal to needle contents (Table 12) except for K which is strongly leached directly after litterfall (section 3.6.3). Initial litter amounts are simulated by assuming that all forest stands in the Netherlands have been planted at the beginning of the 20^{th} century (1910) according to:

$$Am_{lt} = ((1 - fr_{mi}) \cdot k_{lf} \cdot Am_{lv} / kr_{mi}) \cdot (1 - \exp(-kr_{mi} \cdot t)) \tag{161}$$

where t is the time since the forest have been planted (yr). When t is large, the litter amount is in equilibrium with the litterfall. For t a value of 80 has been used (the difference between 1910 and 1990).

Data on the carbonate and Al-hydroxide content are available in a soil information system (Bregt et al., 1986). Data on the total contents of Ca, Mg, K and Na are derived from laboratory analyses on several sandy soils. Average values used in the regional application of RESAM are given in Table 21. Carbonate contents are nihil.

***Table 21** Average values for Al hydroxide - and total base cation contents in A, B and C horizons of acid sandy forest soils of major aerial importance in the Netherlands*

Soil type	Al_{ox} (mmol$_c$ kg^{-1})			Ca+Mg (mmol$_c$ kg^{-1})			K+Na (mmol$_c$ kg^{-1})		
	A	B	C	A	B	C	A	B	C
Leptic podzol	95	185	90	75	70	80	385	375	400
Gleyic podzol	160	220	95	40	55	90	160	295	365
Humic podzol	150	350	115	45	65	105	265	55	370
Anthrosol	95	-	140	100	-	110	540	-	410
Humic gleysol	90	-	30	115	-	130	400	-	470
Albic Arenosol	55	-	65	135	-	75	910	-	400

Table 21 shows that base cation contents in the C horizon are generally quite similar. In the A horizons, the podzolic soils, especially the gleyic and humic podzol, have much lower contents. Furthermore, the total contents of K+Na are much higher than these of Ca+Mg. The total contents of Ca and Mg and of K and Na are generally equal.

Initial cation contents on the adsorption complex are based on field data (Kleijn et al., 1989). Generally, base saturation is less than 10%, whereas Al saturation is more than 60%.

Analogous to SMART, initial anion concentrations in each layer are calculated from the annual atmospheric input and the annual average waterflux in each layer, while the cation concentrations are calculated by combining the charge balance Equation (105) with the various cation exchange cations (125).

5.2.3 Soil properties

Soil properties include the bulk density (ρ), cation exchange capacity (CEC) and sulphate sorption capacity (SSC) (Table 19). These data are all derived by pedotransfer functions.

Values for ρ are related to the organic carbon content according to:

$$\rho = 1 / (a_0 + a_1 \cdot C_{org}) \qquad \text{when } C_{org} \leq 15\% \qquad (162)$$

$$\rho = 0.825 - 0.037 \cdot \log (2 \cdot C_{org}) \qquad \text{when } C_{org} > 15\% \qquad (163)$$

where ρ is the bulk density (gr cm^{-3}) and C_{org} is the organic carbon content (%).

Equation (162) is based on data by Hoekstra and Poelman (1982) for mineral soils and Equation (163) is derived from Van Wallenburg (1988) for peat soils. Values for a_0 and a_1 in acid sandy soils generally range between 0.6 and 0.65 and between 0.04 and 0.06 respectively, depending upon soil type and soil horizon. For the European application with SMART average values of 0.625 and 0.05 have been used independent of soil type.

For the European application the CEC refers to a value measured at pH 6.5. This value is related to both the clay and organic carbon content according to (Helling et al., 1964; Breeuwsma et al., 1986):

$$CEC = 5 \cdot clay + 27.25 \cdot C_{org} \qquad (164)$$

where CEC is the cation exchange capacity ($mmol_c\ kg^{-1}$) and clay is the clay content (%).

The clay content in turn is related to the texture class as given in Table 22.

Table 22 Clay contents (%) of European forest soils as a function of texture class

Class 1	Class 1/2	Class 1/3	Class 2	Class 2/3	Class 3
5	15	25	25	40	50

As with the base saturation given in Table 20, the relation in Table 22 is based on data in FAO (1981) and from Gardiner (1987). The organic carbon content of the various soil types is also derived from these sources. Value range between 0.1% for Arenosols (Oc) and 50% for peat soils (Od). The CEC thus ranges between 25 and 1350 $mmol_c\ kg^{-1}$ (cf Equation (164)).

For the Dutch application with RESAM, the CEC refers to the actual value in the field situation. In acid sandy soils, this value appears to be related to organic carbon only according to (Kleijn et al., 1989):

$$CEC = 15.2 \cdot C_{org} \tag{165}$$

The organic carbon content of the various soil types and soil horizons is derived from the Dutch soil information system (Bregt et al., 1986).

Values for the sulphate sorption capacity, SSC, are related to the content of oxalate extractable Al ($mmol_c\ kg^{-1}$) according to (Johnson and Todd, 1983):

$$SSC = 0.02 \cdot Al_{ox} \tag{166}$$

5.2.4 Soil constants

Soil constants include mineralization, nitrification and denitrification rate constants or fractions, weathering rate constants, dissolution rate constants for $CaCO_3$ and $Al(OH)_3$, cation exchange constants, sulphate adsorption constants and equilibrium constants for the dissociation of CO_2 and dissolution of $CaCO_3$ and $Al(OH)_3$ (Table 19).

Europe

For the application of SMART, mineralization rate constants and dissolution rate constants for $CaCO_3$ and $Al(OH)_3$ are not required.

The nitrification fraction (fr_{ni}) is assumed to vary between 0.75 and 1.0. This is based on NH_4/NO_3 ratios below the rootzone of Dutch forests, which are nearly always less than 0.25 (Heij et al., 1991). The standard value for the nitrification fraction is 1.0. Denitrification fractions are related to soil type on the basis of data given by Steenvoorden (1984) and Breeuwsma et al. (1991) for peat, clay and sandy soils in the Netherlands. Values used are 0.95 for peat soils, 0.80 for clay soils (texture classes 2, 3 and 2/3), 0.5 for sandy soils (texture classes 1 and 1/2) with gleyic features and 0.0 for sandy soils without gleyic features. In deeply drained sandy forest soils, denitrification appears to be negligible (Klemedtson and Svensson, 1988). The ditribution of European forests over peat, non-calcareous clay and sand is 3.7, 28.7 and 58.8%. The remaining part are calcareous soils (8.8%).

Weathering rates required in SMB, START and SMART are zero order constants, i.e. the rate is assumed independent of the amount present. In this context, the term 'weathering' refers to the chemical dissolution of silicate minerals in the soil. Weathering rates are generally estimated from the depletion of base cations in the soil profile, either by chemical analyses of different soil horizons including the parent material or by input output budgets based on hydrochemical monitoring. The former approach gives the average weathering rate over the period of soil formation (e.g. podzolization), whereas the latter gives an estimate of the current weathering rate. Furthermore, weathering models have recently been developed to estimate field weathering rates based on the soil mineralogy (Sverdrup and Warfvinge, 1988a, 1988b). The major input to these models are reaction rate coefficients of minerals and rocks, which are derived from laboratory studies. Using this information, Sverdrup and Warfvinge (1988a, 1988b) established weathering rate classes (approximately 0-500; 500-1000; 1000-1500; 1500-2000 and 2000-2500 and 2500-3000 mol_c ha^{-1} yr^{-1}) on the basis of the mineralogy that controls the weathering rates.

This type of information has been used to assign a weathering rate to European forest soils. The weathering rate is based on the dominant parent material and texture class of the dominant soil unit(s) within each mapping unit. Information regarding the associated soil types is not used. Futhermore, the slope class is considered irrelevant.

The relation used between parent material class, texture class and weathering rate is given in Table 23.

Table 23 Weathering rates used for the various combinations of parent material class and texture class

Parent material class	Weathering rate (mol_c ha^{-1} yr^{-1} m^{-1})					
	1	1/2	1/3	2	2/3	3
Acidic[1)]	250	750	-	1250	1750	-
Intermediate[2)]	750	1250	1750	1750	2250	2750
Basic[3)]	750	1250	-	2250	2750	-

1) Acidic : Sand (stone), gravel, granite, quartzine, gneiss (schist, shale, greywacke, glacial till);
2) Intermediate : Gronodiorite, loess, fluvial and marine sediment (schist, shale, greywacke, glacial till);
3) Basic : Gabbro, Basalt, Dolomite, Volcanic depositis.

Schist, shale, greywacke and glacial till are put between brackets in Table 23. A soil type containing these parent materials can be converted to the acidic or intermediate parent material class, depending on the other parent materials available. Several combinations of parent material class and texture class do not exist below forests. This is indicated with a "-" sign.

Within each class defined before, mean values are used in Table 23. Multiplying this with a soil depth of 0.5 m gives weathering rates of 125, 375, 625, 875, 1125, 1375 $mol_c\ ha^{-1}\ yr^{-1}$.

It has been assumed that texture class has a dominating influence on the weathering rate in the various parent material classes. This is based on a linear relationship between weathering rate and clay content (Sverdrup et al., 1990). However, there is a strong correlation between parent material and texture. Parent material class 1 is mainly associated with texture class 1 and 1/2 while parent material class 2 is mainly correlated with texture class 2 and 2/3.

The assumed dominant parent material class for each soil type on the FAO soil map of Europe below forests is given in Table 24. Two additional classes, i.e. classes 0 (peat) and (4) (calcareous soils; marl, limestone) are also added. The conversion is based on the lithology given in Table 3 of volume V of the FAO soil map of the World (FAO, 1981). An explanation of the various codes is given in Table 5.

Table 24 Proposed conversion between soil type and parent material class

Parent material class				
0	**1**	**2**	**3**	**4**
Od	**A0**	**Bv**	**Th**	**Bc**
Oe	**Bd**	**Ch**	**Tm**	**Be**
	Be	**Cl**	**To**	**Bg**
	Bh	**Gd**	**Tv**	**Bk**
	Dd	**Ge**	**T**	**Ck**
	De	**Gh**		**E**
	Dg	**Gm**		**Hc**
	I[1)]	**Hh**		**Hg**
	Pg	**Hl**		**Jc**
	Ph	**Je**		**Kk**
	Pl	**Kh**		**Lc**
	Po	**Kl**		**Lv**
	Pp	**Li**		**Rc**[2)]
	Qc	**Lg**		**So**
	Ql	**Lo**		**Sm**
	Re	**Mo**		**Vc**
	U	**Vp**		**Xk**
	Wd	**We**		**Xy**

1) Al combinations with I are put in parent material class 1 except for combinations with calcareous soils, that are put in parent material class 4.
2) Rc is differentiated in Rca, Rcb and Rcc.

The weathering rates thus assigned to each soil type by the combination of Table 22 and 23 have been corrected for the effect of temperature according to (Sverdrup, 1990):

$$BC_{we}(T) = BC_{we}(T_o) \cdot e^{(A/To - A/T)} \qquad (167)$$

where BC_{we} (T_o) is the average weathering rate defined in Table 23 for each combination of soil type and texture class, T is the local temperature (K), To is a reference temperature (K) and A is a pre-exponential factor. For A a value of 3600 K has been taken (Sverdrup, 1990). The reference temperature is calculated for each weathering rate class separately. It is the weighted average of the mean annual air temperatures of all soil types in a given weathering rate class. The weighting factor is the percent coverage of the soil type in each grid. Reference temperature (in °C) in the weathering rate classes 1 to 6 equal 4.25, 2.61, 6.52, 8.32, 8.45 and 8.82 respectively. Low temperatures occur for the low weathering rate classes, which mainly occur in Northern Europe. The temperature correction procedure given above implies that the weathering rates given in Table 23 are average values which decrease and increase with a lower and higher temperature.

Cation exchange constants in SMART only refer to the exchange of H and Al with BC. For KH_{ex} a value of $1.5 \cdot 10^5$ is used independent of soil type (De Vries et al., 1989). The value of KAl_{ex} is related to the clay content on the basis of data by Coulter and Talibudeen (1968) and Bache (1974). Values used are 1 for texture class 1, 10 for texture class 1/2 and 1/3, 100 for texture class 2 and 1000 for texture class 2/3 and 3. Values for peat soils (Od and Oe) are taken equal to those for sandy soils.

Values for KCO_2 and KCa_{cb} are taken from the literature (see also De Vries et al., 1989a). For KAl_{ox} a value of 10^8 $(mol\ l^{-1})^{-2}$ is used on the basis of soil solution data in the subsoil of eight Douglas stands (Kleijn et al., 1989) and in 150 Dutch forest stands (De Vries et al.1991b). An exception are the peat soils where KAl_{ox} is put equal to 10^6 $(mol\ l^{-1})^{-2}$ on the basis of literature data (Wood, 1989).

The Netherlands

The mineralization constants needed in the RESAM model are fr_{mi}, $k_{mi,lt}$ and $k_{mi,dr}$ (cf Equation (107) to (109)). Maximum values used, which are reduced as a function of groundwater level and pH, (cf section 3.6.3) are 0.4, 0.05 and 0.7 yr^{-1}. Values are based on a literature survey (De Vries et al., 1990). Values used for DAR and C/N_{mo} (cf Equation (110)) are related to funghi and equal 1.5 and 15 (Janssen, 1983).

Nitrification and denitrification are described different in MACAL and RESAM. MACAL requires nitrification and denitrification fractions. The value of $fr_{ni,in}$ is taken equal to 0.5. This implies that 50% of the NH_4 input is nitrified above the mineral soil, i.e. the litter layer. This is based on experimental data by Tietema et al. (1990). The value of $fr_{ni,rz}$ is assumed to vary between 0.5 and 1.0 based on data on NH_4/NO_3 ratios below Dutch forests (see before). Analogous to the European application, denitrification fractions are related to soil type according to Table 25 (cf Table 7) on the basis of data by Breeuwsma et al. (1991).

Table 25 Denitrification fractions used in the Netherlands for the different soil types

Soil type	Denitrification fraction (-)
Peat	0.95
Clay	0.8
Humic gleysol	0.6
Fluvisol	0.5
Gleyic podzol	0.3
Loess[1)]	0.0

1) Including the remaining sandy soils given in Table 7

Maximum values for the nitrification and denitrification rate constants of the sandy soils included in the RESAM application are derived from calibration on data with respect to nitrification rates (Tietema and Verstraten, 1988; Tietema et al., 1990) and NH_4/NO_3 ratios in the various layers of forest soils (Van Breemen and Verstraten, 1991; Heij et al., 1991). Values used are 30 yr^{-1} and 10 yr^{-1}.

Weathering is also described differently in MACAL and RESAM. MACAL requires zero order weathering rate constants in the rootzone (and subsoil). Weathering rates in the rootzone of acid sandy forest soils in the Netherlands vary mostly between 100 and 400 $mol_c\ ha^{-1}\ yr^{-1}$ (De Vries and Breeuwsma, 1986; De Vries, 1991). Values for the acid sandy soils included in the regional application have been derived on the basis of one-year batch experiments, which have been scaled to field data by dividing them by a factor 50. The value of 50 is based on a comparison of laboratory and field weathering rates estimated by the depletion of base cations in the soil profile (Hootsmans and Van Uffelen, 1991). More information on the discrepancy between weathering rates derived in the field and laboratory is given in Van Grinsven et al. (1988a,b). An overview of weathering rates thus derived is given in Table 26.

Table 26 Weathering rates in the rootzone of acid sandy forest soils of major importance in the Netherlands

Soil type	Weathering rate ($mol_c\ ha^{-1}\ yr^{-1}$)				
	Ca	Mg	K	Na	Total
Leptic podzol[1)]	45	20	75	80	220
Gleyic podzol[1)]	50	95	20	35	200
Humic podzol[1)]	80	165	45	60	350
Anthosol	145	135	70	50	400
Humic gleysol[1)]	40	310	45	75	450
Fluvisol[1)]	60	40	25	35	160
Albic Arenosol[1)]	80	55	40	45	220

1) Values refer to fine textured soils (cf Table 7). Coarse textured variants have a weathering rate which is generally 25% less.

Weathering rates for peat, loess and clay soils are based on literature information (Van Breemen et al., 1984; Weterings, 1989). Values used are 200, 500 and 1000 $mol_c\ ha^{-1}\ yr^{-1}$. More information is given in Hootsmans and Van Uffelen (1991).

RESAM requires first order weathering rate constants (cf Equation (121)). Data have been derived by dividing the weathering rates from the batch experiments described before by the total base cation contents of each sample. Values generally range between 5 10^{-6} and $10^{-4}\ yr^{-1}$ for K and Na and between 2.10^{-5} and $5.10^{-4}\ yr^{-1}$ for Ca and Mg. Weathering rates calculated for the total soil profile are equal to those given in Table 26.

Dissolution rate constants for $Al(OH)_3$, which are needed for the regional application of RESAM, are also based on one-year batch experiments. Values thus derived vary mostly between 10^{-4} and $10^{-8}\ m^3\ kg^{-1}\ yr^{-1}$ for $KElo_1$ and between 5 and 10 for the product of $KElo_2$ and $ctAl_{ox}$ (cf Equation (123)). As with SMART, the value of KAl_{ox} in RESAM is taken at $10^8\ (mol\ l^{-1})^{-2}$. In MACAL KAl_{ox} is calculated as a function of depth (cf Equation (63) and (64)). Values used for α and β are 6.0 and 1.0. This is based on soil solution data at 40 sites and at 4 depths in acid sandy forest soils (Kleijn et al., 1989).

Cation exchange constants for RESAM are derived from the simultaneous measurement of the cations H, Al, Ca, Mg, K, Na and NH_4 on the adsorption complex and in the soil solution of the most important soil types, i.e. all Podzols and the Albic Arenosol (Kleijn et al., 1989). Average values thus derived are given in Table 27.

Table 27 Average cation exchange constants for A, B and C horizons of podzolic soils

Soil horizon	Exchange constants compared to Ca $(mol\ l^{-1})^{2x-2}$					
	H	Al	Mg	K	Na	NH_4
A	40	2.22	0.28	0.30	0.69	168
B	126	16.0	0.36	1.80	2.70	5936
C	39	4.4	0.85	8.05	4.03	5430

Table 27 shows the high affinity of the complex for protons compared to all other monovalent cations. The relative contribution of K, Na and NH_4 on the adsorption complex is very low. For $KSO_{4,ad}$ a value of 1.0 $m^3\ mol_c^{-1}$ has been used in RESAM on the basis of literature information (Singh and Johnson, 1986; Foster et al., 1986).

6.1 Mapping legends

Several kinds of critical load maps can be produced. This includes maps showing for each grid:
- the average critical load;
- the average critical load exceedance;
- the (absolute or relative) area exceeding critical loads.

In order to ensure compatibility with the maps produced in the various European countries (Hettelingh et al., 1991), the map legend for Europe will have five sensitivity classes as shown in Table 28 (after Sverdrup et al., 1990).

Table 28 Legends for mapping average critical loads, critical load exceedances and the area exceeding critical loads

Color code	Raster code	Critical load class ($mol_c\ ha^{-1}\ yr^{-1}$)	Critical load exceedance class ($mol_c\ ha^{-1}\ yr^{-1}$)	Area class exceeding critical loads (%)
red	solid black	< 200	> 1000	> 80
orange	cross hatched	200-500	500-1000	60-80
yellow	diagonally hatched	500-1000	200-500	40-60
light green	dotted	1000-2000	0-200	20-40
dark green	white	2000	< 0	< 20

For the Netherlands a higher degree of resolution might be useful to avoid single color maps. In case, this is needed, the resolution will be made comparible with the sensitivity classes given in Table 28.

Instead of critical loads or critical load exceedances, values can also be calculated for the various chemical criteria such as Al, Al/Ca and pH at a given present deposition rate. Maps showing these data are interesting extra information since critical load calculations imply the choice of a certain critical chemical value, which is rather uncertain for forest soils (section 2). Again, a map legend with five different classes can be used, based on the range in critical chemical values, as shown in Table 29.

Table 29 Legends for mapping chemical criteria

Color code	Raste code	Al ($mol_c\ m^{-3}$)	Al/Ca (mol/mol)	pH
red	solid black	> 1.0	> 5	< 3.5
orange	cross hatched	0.5-1.0	2-5	3.5-4.0
yellow	diagonally hatched	0.2-0.5	1-2	4.0-4.5
light green	dotted	0.1-0.2	0.5-1	4.5-5.0
dark green	white	< 0.1	< 0.5	> 5.0

6.2 Mapping statistics

Within each grid, a range of critical loads and exceedance values exist due to variation in present loads, forest type, soil type, precipitation rate etc. Calculation of an average load can be done by area weighting according to:

$$CL_g = \sum_{i=1}^{n} (A_i \cdot CL_i) / \sum_{i=1}^{n} A_i \tag{167}$$

where CL_g is the average critical load in a grid ($mol_c\ ha^{-1}\ yr^{-1}$), A_i is the area of receptor i (ha), CL_i is the critical load of receptor i ($mol_c\ ha^{-1}\ yr^{-1}$) and n is the number of receptors (forest-soil combinations) in a grid.

In Sverdrup et al. (1990), a weighting procedure is suggested according to:

$$CL_g = \sum_{i=1}^{n} (A_{CLC} / CLCL) / \sum_{i=1}^{n} (1/CLCL) \tag{168}$$

where CLCL is the critical load class limit and A_{CLC} is the area of a critical load class. This implies that the lower critical load classes are more important in the averaging procedure. The same formulas can be applied for calculating the average critical load exceedance or average critical chemical values.

The drawback of average values is that important information on spatial variability can be leveled off. This can be overcome by producing maps that give percentile values of critical loads, critical load exceedances or chemical values e.g. 5%, 50% (median) and 95%. Additional information on the ranges in critical load (exceedances) or chemical values can be given by cumulative frequency distributions for each grid, but this can not be represented on the map itself, unless various grids are clustered (e.g. Kämäri, 1988).

Similarly, histograms can be shown of the absolute frequency (e.g. in km^2) or relative frequency (percentage) of forests in various critical load (exceedance) or chemical criteria classess using the ranges given in Table 28 and 29. The absolute frequency gives insight in both the amount of forest soils and the sensitivity to acid deposition in a grid or cluster of grids.

For the European and national application, maps will be made of the 5, 50 and 95 percentile of critical loads, critical load exceedances and the Al concentration and Al/Ca ratio. Furthermore, frequency diagrams will be made for Europe and the Netherlands as a whole, by plotting the absolute or relative frequency against the various loads and chemical criteria. The latter graphs directly show the impact of the selected critical chemical value on the area exceeding this value, thus visualizing the importance of the chosen criterion.

6.3 Mapping occurence

Grid maps showing critical loads (exceedances) or chemical values are of varying relevance at the different grids due to the variance in the presence of the receptor (forests). Sverdrup et al. (1990) suggested to color or raster a grid square, proportional to the areal occurrence of the receptor in that grid, starting from the middle of the grid. When cartographic presentation allows it, this will be used for the European and Dutch application.

7 SOURCES OF UNCERTAINTY

The uncertainty in load - and critical load exceedance maps is mainly determined by (see also: Alcamo and Bartnicki, 1987; Hettelingh, 1989; Kros et al., 1990):
(1) The uncertainty in the chemical criterium set for the receptor;
(2) The uncertainty in calculation methods (model structure and model implementation);
(3) The uncertainty of data (modelinputs, parameters and initial state of variables due to spatial variability and lack of knowledge.

7.1 Critical chemical levels

The uncertainty in critical chemical levels for a given receptor can be very large. This is especially true for forest (soils) where the range of Al tolerance appears to be very large for different tree species (section 2.2). This directly influences the critical load by the critical alkalinity leaching term (section 3.2.2). This uncertainty, which partly reflects our lack of knowledge regarding the effect of acid deposition on forest vitality, is generally of overwhelming importance for the critical load of forests, especially in areas with a high rainfall (runoff).

This can be illustrated as follows: taking a runoff value of 300 mm yr^{-1} which is a reasonable average value and assuming a critical free Al concentration of 2 mg l^{-1} which corresponds to an alkalinity value of about - 0.3 $mol_c\ m^{-3}$ leads to an critical alkalinity leaching of -900 $mol_c\ ha^{-1}\ yr^{-1}$. However, when a value of 4 mg l^{-1} is assumed this term becomes -900 $mol_c\ ha^{-1}\ yr^{-1}$ has increasing the critical load with 900 $mol_c\ ha^{-1}\ yr^{-1}$. Generally, this is an uncertainty which is higher than the uncertainty in all other parameter values.

The discussion given above does not refer to groundwater and surface waters where the range in critical chemical levels for alkalinity is much lower (section 3.5). For these receptors, the critical load is almost completely determined by the rate of base cation weathering. However, if one wants to avoid depletion of Al-hydroxides, base cation weathering is also of ultimate importance in forest soils (section 3.2.2).

7.2 Calculation methods

Uncertainties in the calculation method relate to the modelstructure and the assumptions that have been made to simplify the "real world". Unlike the uncertainty in chemical values, discussed in the last paragraph, and data, discussed in the next paragraph, it is nearly impossible to quantify the uncertainty due to modeling assumptions. The underlying premise in an uncertainty analysis is that the model structure is correct or at least represent current knowledge adequately. Regarding the models, a large source of uncertainty may occur in various areas due to the occurrence of N-fixation, denitrification or a complex hydrology including seepage or surface runoff. In this context, it is important to note that the use of a one-layer model such as SMB and START will most likely cause an under-prediction of critical acid loads. The annual average water flux in the topsoil is much higher than the precipitation surplus, thus affecting the critical acidity leaching (cf section 3.2.2). In the Netherlands, this difference is approximately 100 to 150 mm yr^{-1}, which causes an increase in acidity leaching of 400 to 600 mol_c ha^{-1} yr^{-1} (De Vries, 1991). The increase in critical load will be of the same order of magnitude, considering that the overall effect of weathering and uptake and nitrogen and base cations is small. The influence of the depth considering on the critical load value will be assessed for Dutch forest soils with the MACAL model.

7.3 Input data

The spatial variability of data is a severe problem when mapping critical loads. It requires insight in the representativeness of data and the validity of extrapolation by vegetation maps, soil maps, geological maps etc. Uncertainty of data is also due to our lack of knowledge and to measurement errors. Unlike spatial variability, which is determined by nature, this source of uncertainty can be decreased by new measurements and improved measurement methods.

Kämäri (1988) and Hettelingh (1989) analyzed the impact of the uncertainty of data, due to spatial variability and lack of knowledge, on the model output of the RAINS model by Monte Carlo analysis, running the model with input data randomly chosen from a given frequency distribution. Such an uncertainty analysis is aimed to be performed both on a European and a National scale, when using the various models i.e. START, MACAL, SMART and RESAM. The model RESAM has already been subjected to such an uncertainty analysis for a single forest soil combination which made it possible to quantify the impact of parameter uncertainty on the uncertainty in model output (Kros et al., 1990). For the SMB model the effect of model input variation on the resulting critical loads, will directly be adressed from Equation (36).

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APPENDIX ON SAMPLING STRATEGIES AND MEASURING TECHNIQUES FOR THE ACQUISITION OF SOIL DATA

An overview of various strategies to gather new soil data is given in Figure 2.

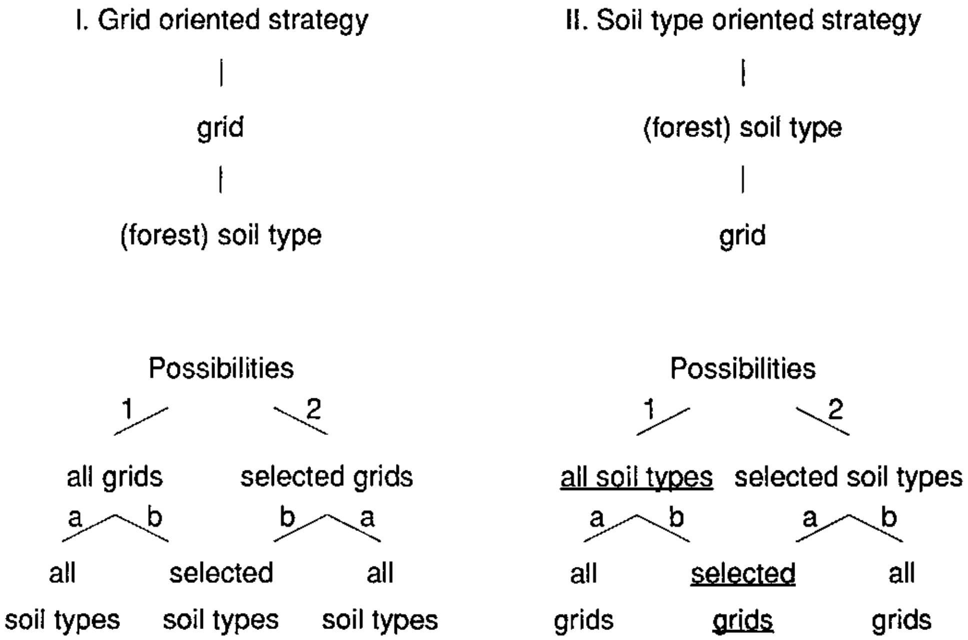

Figure 2 Possible sampling strategies for gathering new soil data

In principle, soil information for each grid can be assessed by sampling all (forest) soil types in all grids (possibility I1a). However, use of this grid oriented strategy leads to a large amount of samples, especially in large countries. This amount might be reduced by sampling only in selected grids with large forested areas (I2a). Furthermore, in both cases one might only sample selected soil types which cover a large area (I1b band I2b). However, the disadventages of the selection of either grids and/or soil types is the possibility of neglecting important (sensitive) soil types.

When the acquisition of soil data is based on a selection of grids and/or soil types it is generally more efficient to use a soil type oriented strategy. Beforehand all (forest) soil types has to be defined and then one can sample these soil types in selected grids only (II1b). In this case there is also the possibility of sampling selected soil types either in all grids (II2a) or in selected grids (II2b). The advantage of sampling all soil types in selected grids (II1b) is that information which is gathered for all soil types can be extrapolated to all grids. However, in order to reduce the amount of soil samples it might be necessary to cluster various soil types into one category. In this respect it is strongly recommeded to use the international FAO classification. By doing so, various soil types in a national classification system are automatically clustered in an FAO soil type and furthermore there is the possibility of international comparison of soil data for similar soil types.

The number of locations that should be sampled to characterize a forest soil can best be related to the area of that soil type. Use of a soil type oriented strategy thus requires information about the area of each soil under forested areas. This can be done by overlaying a forest map and a soil map.

The amount of samples at each location depends upon the spatial variability. The vertical variability is determined by the various soil horizons, and it is strongly recommended to sample each horizon. However, instead of sampling soil horizons there is also the possibility of sampling at fixed intervals. When a mixed sample is taken, in order to reduce the effect of horizontal variability, this is the only practical possibility. As an example, in the context of the ICP on forests, Germany proposes to sample at fixed intervals i.e. 0-5, 5-10, 10-30, 30-60 and 60-90 cm. In the Netherlands 150 forest stands have been sampled at similar depths (0-30 cm, 30-60 cm and 60-100 cm).

An overview of the various measuring methods that are used at the Winand Staring Centre when gathering new soil data is given in Table 30. The data are arranged according to their importance.

Table 30 Measuring techniques for soil data

Variable	Measuring techniques
CEC	Extraction with NH_4-acetate buffered at pH 6.5 or 0.01 N AgTu unbuffered
$frBC_{ac}$[1)]	Extraction with an unbuffered solution e.g., i.e. 0.01 N AgTu
$frAl_{ac}$	ibid
ρ	Sampling a fixed volume and weighting
C_{lt}	Kurmies method (wet digestion)
N_{lt}	Kjeldahl method (wet digestion)
KX_{exc}	Centrifugation of a soil sample in which H, Al, Ca, Mg, K, Na and NH_4 are measured at the adsorption complex and in the soil solution
Ca_{cb}	Wesemael method (weight loss) or Scheibler method (CO_2 evolution)
Al_{ox}	Etraction with NH_4-oxalate buffered at pH 5.5
SSC	Adding SO_4 at a concentration of 200 mg l^{-1} and extraction of the amount at the complex with 0.016 N NaH_2PO_4

1) BC (Ca, Mg, K and Na) can also be measured in a buffered solution

The cation exchange capacity should be measured in a buffered solution in order to standardize to one pH value. There are two possibilities in this respect: use of bariumchloride - tri-ethanolamine ($BaCl_2$-TEA) buffered at pH 8.2 or ammonium - acetate (NH_4OAC) buffered at pH 6.5. Since cation exchange plays a major role in non-calcareous soils with a maximum pH near 6.5, the NH_4OAC method is used. The principle of this method is the measurement of the NH_4 concentration after percolation (columm experiment) or shaking (batch experiment) of a soil sample with a solution of a known NH_4 concentration. The removal of NH_4 is equal to the CEC.

As stated before, it is most important to have information about the base cation fraction at the adsorption complex (par. 3.2.1.2) and this can be measured in the ammonium - acetate percolate. However, in order to measure the Al fraction at the adsorption complex, it is necessary to use an unbuffered solution. In this respect there are various possibilities. At the Winand Staring Centre, the silver-thioreum (AgTu) method is used since the adsorption complex has a very high affinity for Ag. Consequently, the ionic strength of AgTu is only 0.01 N which is in the same order of magnitude as the soil solution concentration. The concentrations of Al, Ca, Mg (and possibly Na and K) in the percolate are measured by atomic emission and atomic adsorption spectrofotometry. The fraction of H at the adsorption complex is derived by substracting the sum of Al, Ca mg, K and Na from the CEC.

The carbonate content of the soil is measured by adding a strong acid (generally HCl) and measuring the weight loss or the CO_2 evolution of the soil sample. The first method (Wesemeal) is fast but less accurate, whereas the second method (Scheibler) is time demanding but more accurate.

The amount of Al in hydroxides which buffer the acid lead can be extracted by various methods e.g. pyrophosphate, ammonium-oxalate and dithionite. The second method is recommended since laboratory experiments indicate that this gives a good indication of the reactive amount of Al.

Carbon and nitrogen in litter can be determined after wet digestion using a mixture of H_2O_2. Total C and total N can than be determined by the Kurmies and Kjeldahl method respectively.

Exchange constants for H and Al versus Ca+Mg (BC) can be derived by measuring the concentrations of H, Ca, Mg and Al in the soil solution which can be extracted by centrifugation. This should be done in samples where the amount at the adsorption complex is also measured. In this way, exchange constants have been derived for different layers (horizons) of acid sandy soils in the Netherlands (Kleijn et al., 1989).

Measurement of SSC requires a laboratory experiment which is rather time demanding (Table 30). The principle of this method is given in Johnson and Todd (1983).