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MAPPING VADEMECUM

J.-P. Hettelingh¹ and W. de Vries²

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1. National Institute of Public Health and Environmental Protection, Coordination Center for Effects, P.O. Box 1, 3720 BA Bilthoven, The Netherlands.

2. The Winand Staring Center for Integrated Land, Soil and Water Research, P.O. Box 125, 6700 AC Wageningen, The Netherlands.

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MAILING LIST

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SUMMARY

The work plan for the implementation of the Convention on Long-Range Transboundary Air Pollution under the UN Economic Commission for Europe (UN ECE) includes the production of maps of critical loads, critical levels, and exceedances as a basis for developing potential abatement strategies for sulphur and nitrogen.

This Vademecum is designed to provide guidance to those responsible for calculating and mapping critical loads, critical levels, and exceedances on a national or regional scale. The chemical criteria and recommended methods to calculate critical loads and levels are described, together with elaboration of the chemical formulas to be used. These procedures were developed from several workshops and studies, which are referenced in the text. In addition, issues related to the practical aspects of producing maps, as well as statistical and uncertainty issues, are discussed.

SAMENVATTING

Het werkplan voor de uitvoering van het VN/ECE verdrag inzake grensoverschrijdende lucht-verontreiniging over lange afstand omvat de produktie van kaarten van kritische waarden, kritische concentraties, en overschrijdingen. Zij vormen de basis voor de ontwikkeling van potentiële strategieën voor het verminderen van emissies van zwavel en stikstof.

Dit Vademecum is opgesteld als een leidraad voor het berekenen en in kaart brengen van kritische belastingen van luchtverontreinigende stoffen, kritische concentraties, en overschrijdingen op nationale of regionale schaal. De chemische criteria en aanbevolen methoden voor het berekenen van kritische belastingen en concentraties zijn beschreven, evenals een nadere uitwerking van de te gebruiken chemische formuleringen. De werkwijzen zijn ontwikkeld op grond van een aantal workshops en onderzoeken, waaraan het rapport refereert. Bovendien worden praktische aspecten van de kaartproductie besproken. Tenslotte wordt aandacht besteed aan onzekerheid in modelberekeningen en de wijze waarop berekeningsresultaten in kaart worden gebracht.

PREFACE

The Executive Body for the Convention on Long-Range Transboundary Air Pollution (LRTAP) of the U.N. Economic Commission for Europe (UN/ECE) has established a Working Group on Strategies (WGS) to assess abatement strategies of sulphur and nitrogen emissions, and a Working Group on Effects (WGE) to assess environmental impacts. The Working Groups are assisted by respectively a Task Force on Integrated Assessment Modelling (TFIAM) and a Task Force on Mapping (TFM). In November 1989 the Executive Body accepted the offer of the Dutch government to establish a Coordination Center for effects-related programs under the Convention, in particular for the mapping of European critical loads and levels. The main current task of the Coordination Center for Effects (CCE) is to assist National Focal Centers (NFCs) in the development of the computational methodology of critical loads and levels including the production of European critical load and level maps. These maps of critical loads and levels are used by the TFIAM for comparison with current deposition and concentrations of acidifying compounds to compute, display and minimize the excess. Furthermore, critical loads of acidity serve as a basis to compute critical loads of sulphur, that is used within the UN/ECE to establish a new sulphur protocol.

Guidelines for the production of the maps of critical loads and levels have been provided in the Mapping Manual and its Annexes that have been provided by the TFM.

This vademecum focusses the manual guidelines, on the practice of producing maps and related efforts. The first draft of the Vademecum appeared in 1990. The vademecum has been updated as a result of discussions held at the Training Sessions organized by the Coordination Center in Bilthoven in 1990 and 1991.

As a result of the guidelines European maps of critical loads were produced and made available in July 1991 (see CCE Technical report no. 1, "Mapping Critical Loads in Europe").

1. GENERAL INTRODUCTION

The aim of this vademecum is to focus on the practice of producing maps for critical loads and levels by:

1. Summarizing the issues related to the computation and presentation of critical loads and levels. This summary is based on, and frequently refers to, the general guidelines that have been formulated in the Mapping Manual and its Annexes produced by the Task Force on Mapping.
2. Including practical guidelines, in cases where the Mapping Manual does not choose between alternatives, for enabling the Coordination Center to present comparable results.
3. Including additional relevant information that is not yet available in the manual and its annexes. This applies to Chapter 5 and to Appendices 1, 2 and 3 of this mapping vademecum, which emerged from discussions at the Training Session.

In this vademecum an effort has been made to provide practical guidelines for producing maps of critical loads and critical levels. Compared to the Mapping Manual the vademecum aimed more at providing pragmatic instructions for producing critical loads maps. Therefore, procedures for producing maps in cases where not all necessary information is available, has been emphasized more than in the Mapping Manual. The aim of this document is to provide support to the improvement of the tools for the evaluation of abatement strategies concerning effects on ecosystems.

As a result of the Training Sessions held in Bilthoven (25-27 June 1990; 14-16 January 1991) this Mapping Vademecum has been adapted based on reports of national experiences. The Training Sessions provided (1) an overview of current gaps in national mapping efforts, (2) an overview of the current experiences elsewhere and (3) an early indication of the results that will be obtained and presented to the Working Groups, the Task Forces and the Executive Body.

Figure 1.1 is a schematic representation of the steps needed to produce European maps describing patterns of (1) critical levels for direct effects of O_3 , SO_2 , NO_2 , and NH_3 on natural vegetation, (2) critical levels for effects on materials, (3) critical loads for acidity, sulphur and nitrogen on freshwater and terrestrial systems, and (4) values by which the mapped patterns of critical levels and critical loads are exceeded by respectively current concentrations and deposition (exceedance maps). Each of the steps represented in Figure 1.1 involves a number of decisions (grid size, critical limits, models) for which guidance is provided by the Mapping Manual. However, the complexity of the methods, available data, available scientific knowledge and infrastructure vary between countries. Depending on the receptor selected, some countries are able to provide very detailed maps, while others are still at the very start of the work.

The emphasis of the mapping exercise was placed on methods and approaches that increase the probability of obtaining European maps rather than multi-country maps, since air, soil and vegetation types have transboundary characteristics. However, various national considerations may lead to differing preferences by which national critical loads and levels are to be assessed. Such preferences may apply to the method applied, the ecosystems addressed, and the results displayed. Therefore, regarding both critical loads and critical levels, two kinds of maps will be made: (1) maps displaying results of different methods and various ecosystems throughout countries, and (2) maps for which one method is applied to one ecosystem used throughout Europe. The first map is the most important map as far as the EB, WGS, WGE, TFIAM and TFM are concerned. The second map is especially important for another task of the CCE: the evaluation of national results in a European framework.

Mapping issues that increase the compatibility mentioned above are emphasized in Chapter 2. Chapters 3 and 4 summarize issues treated in the Mapping Manual regarding critical levels and critical loads, respectively. Chapter 5 treats some aspects of regionalization and uncertainty of the mapping results to be presented.

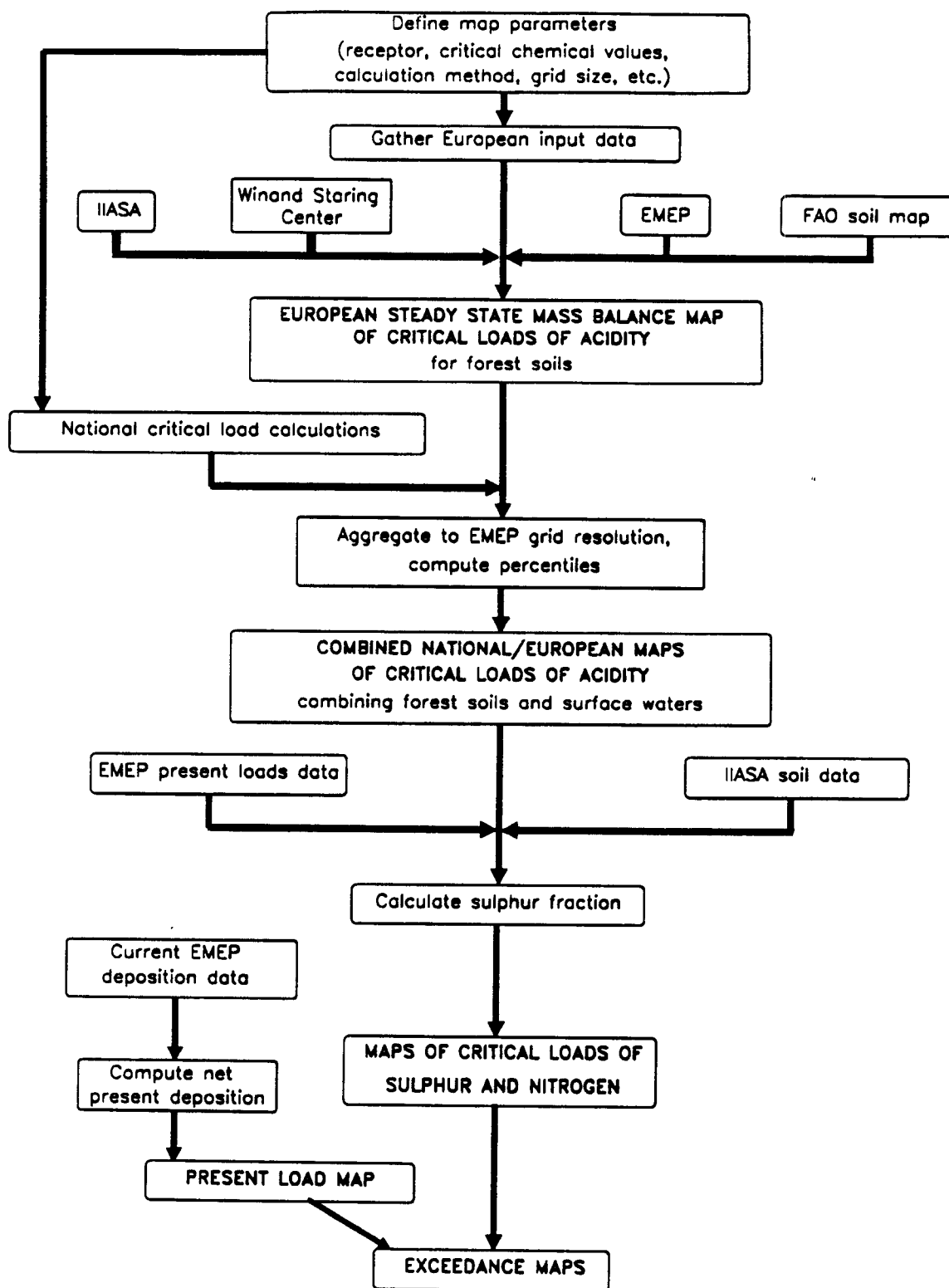


Figure 1.1. Steps in producing critical loads maps for Europe.

2. GENERAL ISSUES OF MAPPING

This chapter provides a tentative overview of general issues related to mapping critical loads, i.e., (1) emission and deposition data that need to be the agreed starting point for the assessing critical loads, critical levels, and their exceedances, (2) map geometry and resolution, and (3) data acquisition, data formats and data presentation.

Specific mapping issues related to critical loads and levels are treated in later chapters.

2.1 Emissions and deposition computations

Computation of effects of O_3 , SO_2 , NO_2 , and NH_3 requires: (1) the estimation of national emissions of sulphur and nitrogen, and (2) the computation and representation of the concentration and deposition of these pollutants.

National emission data used in the mapping exercise should be consistent with those used in the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP). The EMEP-West model computes the deposition of sulphur, nitrogen and ammonia. Recent estimates of the sulphur and nitrogen emissions and deposition can be found in ECE (1989). Concentrations of sulphate, sulphur dioxide, nitrogen dioxide, nitric acid, and ammonia are available from EMEP as well (EMEP, 1990).

Source-receptor matrices (SRM's) have been developed in order to compute, for every EMEP grid cell, the deposition and concentrations of the above-mentioned pollutants, from national emissions. These SRM's are obtained from EMEP's Meteorological Synthesizing Center-West (MSC-W). **A European map of concentrations for ozone is also available; however, a 'SRM for ozone' is not expected to become available soon.** Thus, the evaluation of different control strategies on the current level of ozone, will not be consistent throughout Europe. Several countries may wish to use their own models for the evaluation of abatement strategies. It will depend on (1) the number of countries to produce such national maps and (2) the compatibility between the national ozone level computations, whether a European ozone exceedance map can be produced as result of emission abatement scenario's. Close collaboration with EMEP has been established concerning the evaluation of the compatibility between national ozone level predictions.

Several EMEP-SRM's for SO_2 , NO_2 , and NH_3 have been incorporated in the RAINS model (see also Alcamo *et al.*, 1987, 1990; Hettelingh, 1990, pp. 35-48) of the Transboundary Air Pollution project (TAP) of the International Institute for Applied Systems Analysis (IIASA).

The TFIAM is applying EMEP and models which use EMEP data to assess results of different emission reduction strategies by comparing deposition patterns which result from such strategies. Comparison of critical loads and levels to deposition and concentration patterns will be an important task to evaluate the success of an abatement strategy. Deposition and concentration values predefined by policymakers are termed "*target loads*" and "*target levels*", respectively.

Target loads which are equal to critical loads will, according to the definition of critical loads (see Chapter 4), not cause damage through changes of the chemistry of environmental systems (note that biological effects are not addressed).

2.2 Map geometry and resolution

Information may be geographically represented in two ways, by rasters or polygons. In the Mapping Manual it is recommended that data be represented in rasters. **For this purpose the CCE first provided European maps using the EMEP grid (grid cells of 150 x 150 km²).** It was decided at the first Training Session that national grid square sizes can be chosen according to national requirements. This avoids resolutions which may be considered too coarse, especially for small countries. **Countries should provide the CCE with all the available critical load and level data as well as with the longitude-latitude coordinates at which these critical loads and levels were established.** The final European maps presented to the Executive Body were rastered by the CCE to the EMEP grid in order to (1) guarantee compatibility between country maps, and (2) allow for comparisons with EMEP deposition computations.

Countries may produce detailed national maps to support the research and policy efforts about domestic use of critical loads and levels. It is recommended that such national maps be rastered to 0.25° x 0.125° grids for countries that are smaller or equal to 100,000 km², to 0.5° x 0.25° grids for countries with total area of between 100,000 and 1,000,000 km², and to the above-mentioned 1.0° x 0.5° grids for larger countries (see also Bönsch and Smiatek, 1989).

These maps, and especially the data and their geographical location should also be made available to the Coordination Center to enable integration in a European map rastered according to the EMEP grid. An example of this approach is given in the CCE report (Hettelingh *et al.*, 1991) that has been presented to the Task Forces, Working Groups and the Executive Body.

2.3 Data acquisition

Throughout the Mapping Manual and its Annexes, direct and indirect references are made to monitoring experiments, available site data, or even monitoring experiments to be set up.

The CCE has begun discussions on collaborating with the International Cooperative Programme (ICP) on Integrated Monitoring under the LRTAP Convention. The main tasks of the pilot programme are "the integrated determination and prediction of the state of ecosystems and its changes under the influence of anthropogenic transboundary air pollutants" (UN/ECE, June 1989). A Programme Task Force under the leadership of Sweden reports to the Working Group on Effects and to the EMEP Steering Body about the coordination, planning and evaluation of a Pilot Programme on Integrated Monitoring. Within the framework of this programme a distinction is made, similar to the mapping activities, between National Focal Centers and an Environmental Data Center located in Finland. National Focal Centers for mapping should communicate with NFC's on integrated monitoring in order to assure that the recommendations of the Mapping Manual concerning data monitoring, be consistent and non-overlapping with the work of the pilot programme.

2.4 Data formats and computer systems

The Coordination Center is able to accept files on 3.5 or 5.25 inch diskettes of any density. In addition magnetic tapes (TK70, TK50, 1600 bpi or 6250 bpi) and cartridges (16 and 32 inch) can be processed. For tapes, the number of blocks (bytes), block size (bytes/inch) and type (EBCDIC, ASCII, IBM) should be specified. Files should preferably be transferred as ASCII files, in one of two formats:

- (1) Records describing (a) critical loads, and (b) their geographical longitude, latitude location. This option is appropriate when a large number of particular sites have been sampled and analyzed. Note that each EMEP grid square should be covered by the sampling experiment. A record should first contain the X-coordinate of a value, followed by the Y-coordinate of a value followed by the value itself (i.e., a critical load).

- (2) Records describing (a) a grid square, (b) values (e.g., critical loads) to be represented in that grid square, and (c) the percentage of a grid square to which each value applies. This option is appropriate when the location of values seems less relevant (e.g., one particular value to be applied to a particular forest type covering x% of a particular grid square). A grid square is defined by (1) the longitude and latitude of the lower left-hand corner of that grid square or (2) the EMEP coordinates followed by a value (when records consist of EMEP grid squares). A record should first contain the X-coordinate of a grid square, followed by the Y-coordinate of a grid square, followed by one or more values which apply to the grid square, separated by one or more blanks.

Each of the two formats mentioned above enables the production of cumulative distributions for a grid square. The Coordination Center uses a number of geographical representation and analysis packages; i.e., ARC/INFO, SPANS, ATLAS-GRAPHICS, RAINS and RAINS/GEOMAN. Furthermore, European land use maps will be made available on the RIVM-GIS system, to perform simple overlays for different kinds of maps.

2.5 Geographical data representation

Quantitative geographical information may be represented in many ways like dots, isolines, isopleths, area maps, pseudo-area maps, choropleth and diagram maps (see also Bönsch and Smiatek, 1989). In order to ensure uniformity in geographic presentation, it is decided that the CCE and NFC's should preliminarily restrict to grid shadings to represent the occurrence of particular information (e.g., critical loads). For this purpose the Mapping Manual has defined different boundaries for ranges of critical levels and critical loads.

The information to be mapped often does not coincide with grid cell boundaries (e.g., soil types, deposition, concentrations, plots of forests, crops, urban areas). The choice made to use rastered information, as explained above, makes it necessary to assign the information to grid squares. Each grid square may contain a range of values for each relevant type of information (e.g., ranges of critical loads). The information is displayed by shading patterns which are defined for each interval of value ranges of the information as given in Table 2.1. In some cases not all the information is needed for all the grid squares of Europe, e.g., material damage (restricted to urban areas). In such cases the representation of the information should be restricted to relevant grid squares. Class boundaries depend on the data to be mapped and are subsequently given in the next chapters.

Table 2.1. Shading patterns for each information class.¹

Class	Black & White	Color
Very important	solid black	red (magenta)
Important	dark grey	orange (red)
Less important	grey	yellow (yellow)
Lesser important	light grey	green (light blue)
Least important	solid white	blue (dark blue)

The Mapping Manual makes a distinction between primary and secondary maps. Primary maps display loads or levels. Secondary maps contain the kind of background information (e.g., forested areas, urban areas, lake areas) which is important for the interpretation of maps displaying loads or levels. It is recommended to avoid maps containing too much information by overlaying several maps onto a single

1. Actual class boundaries depend on the issues to be classified and will be made explicit in the following sections.

display. Clipping methods (showing critical levels for materials only in grid squares where it is relevant) will increase the comprehensiveness of the displayed information.

There are two possibilities to geographically classify the information on critical levels or loads, depending on data and model results:

1. A method, based on a weighing scheme, is proposed in the manual annex (Sverdrup *et al.*, 1990) in order to define to which class the grid square information on critical loads should be allocated. The weighing method leads to: (a) one number only and (b) a shading pattern displaying the occurrence (i.e., part of the grid square is shaded) of the information within a grid square. The map displaying a shading pattern established according to the weighing scheme described above is likely to be difficult to read due to different sizes of shaded grid squares and be very sensitive to the chosen weighing scheme.
2. As an alternative, it is recommended to produce a cumulative distribution of the occurrence of each type of information within a grid square, in order to be flexible about the choice of the representation of any percentile. **Provided the data is transferred as specified in Section 2.4, the CCE can produce cumulative distributions for each of the EMEP grid squares.**

A particular percentile, rather than cumulative distributions per grid square, are presented on the final maps. At the second training session it was decided to produce three maps of critical loads: the 1, 5, and 50 percentiles of critical loads in each grid square. The grid squares will be shaded according to the value range to which the 1%, 5%, or 50% critical load belongs. The application of multiple percentiles will enable (1) verification of the extent by which information is biased by the chosen statistic (i.e., percentile), and (2) quantification of shifts in class occurrences in a grid square due to results of abatement strategies.

It is assumed that information can be assigned to grid squares. However, in the case that information is based on (rough) samples (e.g., number of lakes in an area, a few measurements of needed parameters in a few areas or grid squares) not all grid squares are necessarily covered by a set of data. In such cases it will also be possible to produce statistical information (density functions, cumulative distributions) for (functional) regions for which the samples are assumed to be representative. The method of quantifying percentiles, described above, still applies with the only difference that the map will consist of shaded regions instead of shaded grid squares.

One should be aware that a display of percentiles for large regions is less accurate than for small regions (i.e., grid squares). A 95% critical load in the region of Europe is likely to correspond to a high value, even if this value hardly occurs. A 95% critical load in a small grid square will correspond to a lower critical load value. In summary it should be noted that the display of a particular percentile in the EMEP grid is likely to show an area of sensitive ecosystems which is smaller than the area displayed for the same percentile in the 1° x 0.5° longitude/latitude grid.

Other issues related uncertainty and to regional (model) applications can be found in Chapter 5.

3. CRITICAL LEVELS

This chapter presents a short overview of preliminary criteria, methods, data and mapping issues that need to be taken into account for the mapping of critical levels for effects on vegetation and materials.

3.1 Vegetation

3.1.1 Criteria

The definition of critical air concentrations of chemical compounds such as O₃, SO₂, NO₂, and NH₃ with respect to direct effects depends on the time of exposure. Critical concentrations decrease when the exposure time increases. Therefore the Mapping Manual (Chapter 3) defines a relation between the compound, the exposure duration and related definitions of critical levels. The information is summarized in Table 3.1. NH₃ levels are excluded since confirmation about a critical level has not yet been defined.

Table 3.1. Preliminary critical levels for O₃, SO₂¹, and NO₂¹.

Compound	Land use	Critical level (µg/m ³)	Time period
O ₃	all	50	7 hr. mean ²
SO ₂	forest ³	20	annual mean
	natural vegetation	20	annual mean
	crops	30	annual mean
NO ₂	all	30	annual mean

1. Source: Mapping Manual Chapter 3, tables 3.2, 3.3, 3.4.
2. Growing season.
3. Mapping of effects to forest should have priority.

3.1.2 Methods

Calculating exceedances of critical levels consists of comparing actual air concentrations and critical levels for which tables are presented in the Mapping Manual (see Chapter 3), which have been summarized in Table 3.1. The manual distinguishes between two approaches. The first approach, ("Level 1"), distinguishes only major land use types such as forests, crops and natural vegetation, as sensitive receptors. In the Level 2 approach, individual species are taken into account. Although some countries may make progress with Level 2 it is unlikely that a compatible result between all countries will be achieved in the first maps. Specific national Level 2 results could be included in a CCE report on critical levels to the Working Groups, Task Forces and EB.

3.1.3 Data

Land use data are generally available (Mapping Manual, Chapter 3, table 3.5). NFC's are urged to provide the CCE with national land use data to allow the extension of the GIS database of the CCE.

Data on ambient concentrations should be consistent with EMEP as much as possible. As described in Section 2.4, the CCE can assess cumulative distributions from data as (1) critical levels including their location, or (2) grid square locations and a set of critical levels assigned to each grid square.

An excess of critical levels will be computed for each EMEP grid square by subtracting ambient concentrations of a chemical compound from a critical level in each EMEP grid square. National concentrations, based on model runs and monitoring results, should preferably be used. An overview of existing models is provided by Derwent and Ries (1989).

3.1.4 Mapping issues

The European maps of (a percentile of) critical levels for different chemical compounds will display a concentration level in each EMEP grid square.

Shading class limits should be obtained by subdivision of the applicable data range into 5 sub-ranges, as mentioned in Section 2.6. Table 3.2 displays the shading limits for (1) annual SO₂ levels on forests and natural vegetation, (2) annual mean SO₂ levels on crops, (3) annual mean NO₂ levels on all land use types and (4) O₃ levels for the 7-hour mean averaged over the growing season.

Table 3.2. Proposed legend for the classifications¹ of average SO₂, NO₂ and O₃ levels. (µg/m³).

SO ₂		NO ₂	O ₃
Forests ²	Crops	All	Crops ³
> 20	> 30	> 30	> 50
15 - 19.9	23 - 29.9	23 - 29.9	37 - 49.9
10 - 14.9	16 - 22.9	16 - 22.9	24 - 36.9
5 - 9.9	9 - 15.9	9 - 15.9	11 - 23.9
< 5	< 9	< 9	< 11

1. Shading patterns are described in Table 2.1.
2. Including natural vegetation.
3. 7-hour mean (9.00-16.00) averaged over the growing season (1 April - 30 September).

A final decision about the choice of particular percentiles of critical levels to be displayed will be investigated as the first results become available.

3.2 Materials

3.2.1 Criteria

Critical levels for effects on materials are not exclusively dependent on the concentration of gaseous pollutants, i.e., O₃, SO₂, NO₂, and NH₃, as damage to materials occurs due to interaction between pollutants, climatic factors (e.g., temperature, humidity), particulate and other chemical compounds (e.g., Cl⁻, SO₄²⁻, H⁺). Furthermore, the extent to which damage occurs varies for different materials (see Mapping Manual, Chapter 5). Therefore, it is not possible to define critical levels for the effects of pollutants on materials below which no deterioration occurs.

However, the effect of particular exposure limits could be expressed in terms of changes in the expected lifetime of a material. Target levels could be defined such that acceptable lifetimes for materials are reached (Mapping Manual, Chapter 5). In the manual a distinction is made between (1) cultural heritage and (2) modern objects. For cultural heritage, the stock at risk needs to be determined on a national basis. For modern objects, acceptable lifetimes should be as long as their economic lifetimes.

3.2.2 Methods

No specific methods are suggested in the manual. The major problem is the estimation of the stock at risk. In Gosseling *et al.* (1989) the damage to so-called Baumberger stone (Luckat, 1981) is used as an approximation to all natural stone monuments in the Netherlands (Gosseling *et al.*, 1989, p. 26). Another theoretical approach, to define the half-time of material and the price of the increasing scarcity of a material as a consequence of damage, can be found in Benarie (1989).

The CCE proposes at present to overlay maps of (1) concentrations of aggressive compounds (i.e., SO₂, NO₂), (2) climatic data (i.e., temperature, rainfall) (3) infrastructure (i.e., cities, roads, industrial areas).

3.2.3 Data

Much of the success for mapping European material data will depend on national research results. Concerning concentration data, an approach similar to critical levels for effects on vegetation could be adopted (see Section 3.1).

3.2.4 Mapping issues

Maps should concentrate on those areas where material is allocated, i.e., urban and industrial areas. A grid square size is recommended in the Mapping Manual that does not exceed 50 x 50 km squares.

The Working Group on Effects has implemented an International Cooperative Programme on Effects on Materials, including Historic and Cultural Monuments (UN/ECE, 24 July 1989). A preliminary result of exposure surveys is now available. An overview of the sites where exposure data is collected, has been made by the Main Research Center at the Swedish Corrosion Institute in August 1989. The Coordination Center will establish necessary contacts to remain updated with the results of this programme.

Meanwhile, as a first step maps of O₃, SO₂, and NO₂ concentrations could be overlaid with maps displaying urban and industrial areas. If possible, frequency distributions of the occurrence of material objects (e.g., houses, roads, monuments, machines, means of transport) in each relevant grid square (i.e., those containing urban or industrial areas), or region should be provided as well.

4. CRITICAL LOADS

The Mapping Manual has an extensive and detailed Annex on critical loads mapping (Sverdrup *et al.*, 1990). In this chapter a summary of the critical loads approach is presented.

4.1 Criteria

In the assessment of critical loads for acidity on fresh waters and terrestrial systems a distinction is needed between the effects of acidification due to both sulphur and nitrogen deposition and eutrophication due to nitrogen deposition only. Therefore both a *critical acid load* and a *critical nitrogen load* should be computed for soils. With respect to freshwaters only a critical acid load is computed, as it is assumed that nitrogen is completely taken up by the vegetation.

In order to compute critical acid loads it is necessary to determine critical alkalinity limits. These limits can be derived from critical limits for pH and aluminum as follows (see also de Vries, 1991).

$$[\text{Alk}] = [\text{HCO}_3^-] + [\text{RCOO}^-] - [\text{H}^+] - [\text{Al}^{3+}] \quad (1)$$

where:

- [Alk] = alkalinity (acid neutralizing capacity) ($\text{kmol}_c \text{ m}^{-3}$)
- $[\text{HCO}_3^-]$ = concentration of bicarbonate ($\text{kmol}_c \text{ m}^{-3}$)
- $[\text{RCOO}^-]$ = concentration of organic anions ($\text{kmol}_c \text{ m}^{-3}$)
- $[\text{H}^+]$ = concentration of H^+ ions ($\text{kmol}_c \text{ m}^{-3}$)
- $[\text{Al}^{3+}]$ = concentration of aluminum ions ($\text{kmol}_c \text{ m}^{-3}$)

In soils the critical pH is such that the concentration of bicarbonates ($[\text{HCO}_3^-]$) can be neglected. Organic anions ($[\text{RCOO}^-]$) can also be neglected, assuming that RCOO^- is completely associated with aluminum. This implies that aluminum only refers to toxic inorganic species. Assuming gibbsite equilibrium, the critical pH can be computed from a given critical aluminum concentration as follows:

$$\text{pH} = (\log K_{\text{gibb}} - \log[\text{Al}]) / 3 \quad (2)$$

where:

- pH = unit for the measurement of $[\text{H}^+]$
- K_{gibb} = gibbsite equilibrium constant ($\text{kmol}_c \text{ m}^{-3}$)
- [Al] = aluminum concentration ($\text{kmol}_c \text{ m}^{-3}$)

From equations 1 and 2, neglecting $[\text{HCO}_3^-]$ and $[\text{RCOO}^-]$, a critical alkalinity concentration can be computed. For freshwaters the aluminum concentration may be neglected for computing critical alkalinity. However bicarbonates are included since the critical pH is much higher than in forest soils. Bicarbonates can be computed from the critical pH according to:

$$[\text{HCO}_3^-] = K_1 \cdot K_H \cdot p\text{CO}_2 / [\text{H}^+] \quad (3)$$

where:

- K_1 = first acidity constant ($\text{kmol}_c \text{ m}^{-3}$)
- K_H = Henry's law constant ($\text{kmol}_c \text{ m}^{-3} \text{ atm}^{-1}$)
- $p\text{CO}_2$ = partial pressure of CO_2 (atm)
- $[\text{H}^+]$ = concentration of H^+ ions ($\text{kmol}_c \text{ m}^{-3}$)

Substitution of equations 2 and 3 into equation 1, neglecting [Al], leads to the computation of critical alkalinity in freshwaters.

In order to derive a critical nitrogen load it is necessary to determine critical nitrate concentrations. Table 4.1 gives an overview of average critical values that can be obtained from the literature (Al, Al/Ca, NO₃) and by equations 1 through 3 (pH, Alk). The average values have been computed from values that can be found in Sverdrup *et al.* (1990) and de Vries (1991). It should be noted that the values, mentioned above, may be large especially for [Al] and Al/Ca in forest soils.

Table 4.1. Acceptable limits for chemical compounds in forest soils and freshwater systems.

Compound (unit)	Forest soil	Freshwater	Groundwater
[Al] mol _c m ⁻³	0.2	0.003	0.02
Al/Ca mol mol ₁	1 ¹	-	-
pH	4 ²	(5.3,6.0) ³	6.0
[Alk] mol _c m ⁻³	-0.3 ²	(0.02,0.08) ³	0.14
[NO ₃] mol _c m ⁻³	-	-	0.8

1. Using an equivalent ratio, a value of 1.5 mol_c mol_c⁻¹ is used.
2. Assuming log K_{gibb} of 8.0 and [Al]= 0.2. (A pH of 4 corresponds to 0.1 mol_c m⁻³ of H⁺). See equations 10 to 13 for a more general computation of acceptable alkalinity.
3. A pH of 6.0 relates to peak flow situations and is associated with [Alk] = 0.08 mol_c m⁻³.

4.2 Methods

In the assessment of critical loads for forest soils and freshwaters the understanding of chemical processes in soils and in surface waters is increased by modeling. In the Mapping Manual a distinction is made between (1) steady-state modeling (Level I), divided in a water chemistry method and a mass balance method, and (2) dynamic modeling (Level II).

The steady-state modeling approach assumes a time-independent steady state of chemical interaction involving an equilibrium between the production and consumption of acidic compounds. The dynamic approach can be used to assess critical loads in relation to time-dependent processes thus enabling the assessment of the current and future affects of abatement strategies.

Another, more qualitative approach (the "Level 0" approach) which has been applied in the United Kingdom, is presented in Appendix 1.

4.2.1 Steady state methods for surface waters

The key equation for the steady state mass balance computation of critical loads in **freshwaters**, according to the *Water Chemistry Method* (Henriksen *et al.*, 1988; see also Sverdrup *et al.*, 1990) is as follows:

$$CL(Ac) = BC_0^* - BC_{td}^* - Q \cdot [Alk]_{crit} \quad (4)$$

where:

CL(Ac) = critical load of acidity in freshwater (mol_c ha⁻¹ yr⁻¹)

BC₀^{*} = product of base cation concentration and runoff (mol_c ha⁻¹ yr⁻¹)

BC_{td}^{*} = the seasalt-corrected deposition of base cations (mol_c ha⁻¹ yr⁻¹)

Q^l = runoff ($\text{m}^3 \text{ ha}^{-1} \text{ yr}^{-1}$)
 $[\text{Alk}]_{\text{crit}}$ = critical alkalinity concentration ($\text{mol}_c \text{ m}^{-3}$)

The following relationship holds in an equilibrium situation:

$$\text{BC}_0^* = \text{BC}_d^* + \text{BC}_w - \text{BC}_u \quad (5)$$

where:

BC_w = weathering of base cations (Ca, Mg, K, Na) ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 BC_u = net uptake of base cations ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)

Substitution of equation 5 into equation 4 yields:

$$\text{CL}(\text{Ac}) = \text{BC}_w - \text{BC}_u - Q \cdot [\text{Alk}]_{\text{crit}} \quad (6)$$

Critical loads for nitrogen are not computed in the present form of the Water Chemistry Method.

4.2.2 Steady state methods for soils

The *Steady State Mass Balance (SSMB) Method* may be used for the computation of a critical load of acidity for soils, groundwater and surface water. In the following the computation of critical loads in forest soils is emphasized. A distinction is made between the critical load for (a) potential acidity, (b) actual acidity, and (c) free acidity.

(a) Critical load for potential acidity:

The following key equation is applied (de Vries, 1991; Sverdrup *et al.*, 1990, chapter 2):

$$\text{CL}(\text{Ac}_{\text{pot}}) = \text{BC}_w - \text{BC}_u + N_u + N_{i(\text{acc})} - \text{ANC}_{l(\text{acc})} \quad (7)$$

where:

$\text{CL}(\text{Ac}_{\text{pot}})$ = critical load of potential acidity ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 BC_w = weathering of base cations (Ca, Mg, K, Na) ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 BC_u = net uptake of base cations ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 $N_{i(\text{acc})}$ = acceptable immobilization of nitrogen ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 N_u = net uptake of nitrogen ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 $\text{ANC}_{l(\text{acc})}$ = acceptable alkalinity leaching (see equation 10)

Equation 7 only accounts for deposition, weathering, net uptake of base cations and net uptake of nitrogen in one compartment. However, when in addition litterfall and nutrient uptake within the root zone is assumed, then a multi-layer model such as MACAL or PROFILE should be used (de Vries, 1991; Sverdrup and Warfvinge, 1988).

(b) Critical load for actual acidity:

1. Q can be defined as the flow rate through a system. If the system consists of watersheds, then Q is defined as precipitation minus evapotranspiration. If the system consists of soils, then the flow rate should be defined as precipitation minus evapotranspiration minus surface runoff, the result of which is referred to as precipitation surplus (PS). Throughout this report Q will be used even when soils are considered.

The effects of base cation uptake, nitrogen uptake and nitrogen immobilization should not be included when calculating the critical load for actual acidity produced in the soil system. The idea behind the exclusion of BC_u , N_u and N_i is that critical loads should be computed independent of land use changes which may affect BC_u , N_u and N_i . Exclusion of these three elements from equation 7 leads to the following key equation for the computation of the critical load for actual acidity [$CL(Ac)$]:

$$CL(Ac) = BC_w - ANC_i \quad (8)$$

(c) Critical load for free acidity:

The Mapping Manual also describes an expression for the computation of critical loads which includes ammonia as a base cation. Thus, from equation 7, the following key equation for the computation of free acidity [$CL(Ac_{free})$] is derived:

$$CL(Ac_{free}) = CL(Ac_{pot}) - 2 \cdot NH_{4,d} \quad (9)$$

Equation 9 includes an atmospheric variable, i.e., the deposition of ammonia, and hence leads to a critical load which is not merely based on ecosystem characteristics. Consequently, for the mapping exercise, use of the critical load of actual acidity is favored, as a result of discussions during the January 1991 Training Session.

More information on the relationship between potential acidity (equation 7), actual acidity (equation 8) and free acidity (equation 9) is given in Appendix 2, which also includes a discussion on the derivation of critical loads for sulphur and nitrogen. The reason for a sulphur critical load came about during the July 1990 meeting of the Working Group on Abatement Strategies, at which representatives asked whether maps on acidity and potential acidity could be used for the model assessments needed to reach a new sulphur protocol.

Acceptable alkalinity leaching:

Acceptable alkalinity leaching, $ANC_{l(acc)}$, is important in each of the three equations (7, 8, and 9), and is defined as follows for acid forest soils:

$$ANC_{l(acc)} = -H_{l(acc)} - Al_{l(acc)} \quad (10)$$

where:

$$H_{l(acc)} = Q \cdot [H]_{acc} \quad (11)$$

and:

$$Al_{l(crit)} = \begin{cases} Q \cdot [Al]_{crit} & \text{(the Al criterion), or} \\ R(Al/Ca)_{crit} \cdot (BC_{dt}^* + BC_w - BC_u) & \text{(the Al/Ca criterion)} \end{cases} \quad (12)$$

where:

$H_{l(acc)}$ = acceptable hydrogen leaching ($\text{mol}_e \text{ ha}^{-1} \text{ yr}^{-1}$)
 $Al_{l(acc)}$ = acceptable aluminum leaching ($\text{mol}_e \text{ ha}^{-1} \text{ yr}^{-1}$)
 $[H]_{acc}$ = acceptable hydrogen concentration ($0.1 \text{ mol}_e \text{ m}^{-3}$)¹

1. $[H]_{acc}$ is computed from: $[H]_{acc} = ([Al]_{acc}/K_{gibb})^{1/3}$ (11a)
 where:

K_{gibb} = gibbsite solubility constant ($3 \cdot 10^2 (\text{mol m}^{-3})^{-2}$)

$[Al]_{acc}$ = acceptable aluminum concentration ($0.2 \text{ mol}_c \text{ m}^{-3}$)

$R(Al/Ca)_{acc}$ = acceptable equivalent Al/Ca ratio ($1.5 \text{ mol}_c \text{ mol}^{-1}$)

BC'_{dt} = total seasalt-corrected deposition of base cations ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)

Besides the Al and Ca/Al criterion, a third criterion, the aluminum depletion criterion, may be distinguished as follows: $Al_{l(acc)} = r \cdot BC_w$, where r is the stoichiometric weathering ratio of Al to BC_w . However, the Al/Ca and Al criteria are considered more relevant from an ecosystem point of view.

Substitution of equations 10 to 13 into equation 8 leads to the following two equations for the critical load of actual acidity:

$$CL(Ac) = \begin{cases} BC_w + 0.09 \cdot Q + 0.2 \cdot Q & \text{if equation 12 is used} \\ BC_w + 0.09 \cdot Q + 1.5 \cdot (BC'_{td} + BC_w - BC_u) & \text{if equation 13 is used} \end{cases} \quad (14)$$

(15)

The lower of the two values respectively calculated by equations 14 and 15 is to be used.

Note, from equations 4 to 15, that the water flux (runoff) may have a considerable influence on the critical load computation, which varies for different receptors. Critical alkalinity in forest soils is negative (see Table 4.1). A consequence of a negative critical alkalinity is that an increasing water flux will yield a higher critical load. The opposite is true for groundwater and surface water where the critical alkalinity is positive. The reason is that an increasing water flux will increase the dilution of aluminum. However, since also calcium is diluted the Al/Ca ratio is not affected. Thus it is recommended to apply both the Al and Ca/Al criteria (see Sverdrup *et al.*, 1990) and take the minimum critical load from equations 14 and 15.

One of the differences between the Level 0 approach mentioned earlier (see Appendix 1) and the application of the steady state mass balance is that the level 0 approach will yield a decreasing critical load as the water flux increases (see Nilsson and Grennfelt, 1988; Appendix 1, Table 3). The reason is that an increasing water flux will increase the base cation depletion. However, in the steady state approach base cation depletion may be considered a dynamic process which is not taken into account. (The same applies to sulphate adsorption capacity mentioned in Table A1.3 of Appendix 1.)

It is necessary to investigate the consequences of this difference in critical load estimation more thoroughly especially since a good understanding is needed for a suitable (dynamic) assessment of acid (sulphur and nitrogen) abatement scenarios. The importance of the temporal evolution of (critical) chemical parameters as a result of abatement strategies may become important especially in areas where the estimated critical load is exceeded.

Critical load of nitrogen:

The critical load of nitrogen as a nutrient becomes:

$$CL(NN) = N_u + N_{i(acc)} + Q \cdot [NO_3]_{acc} \quad (16)$$

$[Al]_{acc} = Al_{l(acc)}/Q$ (computed from equations 12 or 13).

When assuming $[Al]_{acc} = 0.2 \text{ mol}_c \text{ m}^{-3}$ (see Table 4.1) and K_{gibb} as above, it follows from equation (11a) that $[H]_{acc} = 0.09 \text{ mol m}^{-3}$, which in Table 4.1 has been rounded to 0.1 (pH=4 in Table 4.1).

In equations 14 and 15 the value of 0.09 has been substituted for $[H]_{acc}$. National Focal Centers may of course apply equation 11a to obtain other values.

where:

CL(NN) = critical load of nitrogen as a nutrient ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 N_u = net uptake of nitrogen ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 $N_{i(\text{acc})}$ = acceptable immobilization of nitrogen ($\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$)
 Q = runoff ($\text{m}^3 \text{ ha}^{-1} \text{ yr}^{-1}$)
 $[\text{NO}_3]_{\text{acc}}$ = acceptable nitrate concentration ($\text{mol}_c \text{ m}^{-3}$)

Values for $[\text{Alk}]_{\text{acc}}$ and $[\text{NO}_3]_{\text{acc}}$ are given in Table 4.1. Values for the other variables are dependent on receptor characteristics.

In summary, the steady state mass balance approach is by far the simplest method to assess critical loads and has rather limited data requirements (see Section 4.3, "Data"). In cases where more detailed survey data are available it is recommended to use the PROFILE model (Sverdrup and Warfvinge, 1988; Warfvinge, 1988) or MACAL (de Vries, 1991).

4.2.3 Dynamic models

The dynamic modeling method involves the application of models in which time-dependent chemical processes in soils and freshwaters are simulated. The main purpose of the dynamic modeling approach is to predict the time period that elapses before a critical (acceptable) chemical value is reached. Dynamic models enable the computation of chemical criteria (i.e., pH, Al concentration, molar Al/Ca ratio, and NO_3) and of critical load exceedances, through time.

It is expected that a long-term exceedance of critical loads, computed using the steady state mass balance approach, will at some point in time, lead to a violation of critical chemical values presented in Table 4.1. Thus it becomes possible to evaluate abatement strategies with respect to (1) the time horizon needed to reach particular target chemical values, or (2) the time horizon that remains before a critical chemical value is reached. As a result, the evaluation of abatement strategies need not be limited to the question whether a critical load is exceeded, without inspecting the extent of an exceedance. An inspection of the time development of critical chemical values resulting from a reduction of a critical load exceedance will provide better understanding of the benefits of such abatement strategies.

In the Mapping Manual several dynamic models are mentioned. Concerning regional soil acidification assessments on a European scale, it is recommended to use the Simulation Model for Acidification's Regional Trends (SMART; de Vries *et al.*, 1989). This model estimates soil acidification throughout Europe. SMART is now being linked to the Energy, Emission and Deposition Modules of RAINS. With respect to watershed acidification it is recommended to use the MAGIC model (Cosby *et al.*, 1985a, 1985b) or SMART.

4.3 Data

Data requirements differ from model to model, and are well described in the Mapping Manual. Data requirements for the Steady State Mass Balance Approach and for PROFILE and MACAL are given in the Table 4.5 of the manual. Data requirements for the water chemistry approach are described in manual section 4.2.2.1, and data requirements for SMART and for MAGIC are described in Table 4.8 of the manual.

An overview of data acquisition procedures and suggested default values for the most important parameters in several dynamic models is given in Sverdrup *et al.* (1990). This background document also includes a presentation of different methods, including the application of PROFILE, to estimate the weathering rate. When the necessary input data for PROFILE are available, PROFILE should be used for

the estimation of weathering rates. However, the estimation of the weathering rate may require information on the mineralogy, which reaches a level of detail that may be too demanding for some countries to allow for first results to be obtained by early next year. Therefore, a simple estimation procedure for the derivation of weathering rates from soil type information is presented in Appendix 3. A more detailed overview of this estimation procedure, focused on the SMART model, is presented in de Vries (1991).

Data bases for applying the steady state mass balance model at a spatial resolution of $1^\circ \times 0.5^\circ$ longitude-latitude are available at IIASA. However, most inputs are estimated or empirically derived (e.g., the weathering rate) and many of the data requirements need further surveys throughout Europe. Validation of the empirically derived relations and other assumptions may need more research time. Therefore, in the representation of the results, it is necessary to take uncertainty into account by providing ranges of model results rather than one deterministic outcome (See Chapters 1 and 5).

4.4 Mapping Issues

The European map of critical loads will ideally consist of (1) a map of shaded grid squares, and (2) an overview of cumulative distributions of critical loads for each grid. This format has been used by Kuylenstierna and Chadwick (1989) to produce a first qualitative map of critical loads over Europe which was successfully used for the evaluation of abatement strategies.

It was decided at the first Training Session that the critical load map of Europe should be related to the percentage of the grid square area (or percentage of data points within a grid square) being displayed. It was decided at the second training session that the 5-percentile and 50-percentile European map of critical loads will be produced, displaying the range of critical loads which is covering respectively 5% and 50% of the grid square or data points (lakes) within the grid square (see Section 2.5). Without getting into too many statistical details it can be noted that a 5 percentile critical load represents the deposition at which, 95% of -broadly speaking- the grid cell area is protected. A complication is introduced when a mixture of data points and grid square areas occur in one grid square (e.g., a grid square containing critical loads for forests, lakes and groundwater).

Uncertainty analysis (Chapter 5) may be a suitable method in cases where NFC's have doubts on the accuracy of the data used in each grid square. Uncertainty analysis generally consists of applying a Monte Carlo method on the steady state mass balance equation (equations 13 and 14) for computing critical loads of acidity. In the latter application, ranges of input values for the right hand variables need to be assumed. Inputs are generated (see Chapter 5) for the steady state mass balance equation by repeated sampling from these ranges. This will yield cumulative distributions of critical loads for the grid squares where the method is applied.

Finally, using the shading patterns defined in Chapter 2 (Table 2.1), ranges (Table 4.2) are distinguished for (1) critical loads and (2) deposition above present critical loads.

Table 4.2. Ranges of critical loads and critical load exceedance.

Critical load range ($\text{kmol}_e \text{ ha}^{-1} \text{ yr}^{-1}$)	Exceedance of critical load ($\text{kmol}_e \text{ ha}^{-1} \text{ yr}^{-1}$)
0.00 - 0.20	< 0.00
0.21 - 0.50	0.00 - 0.20
0.51 - 1.00	0.21 - 0.50
1.01 - 2.00	0.51 - 1.00
> 2.00	1.01 - 2.00
	> 2.00

5. UNCERTAINTY AND SPATIAL GENERALIZATION

5.1 Introduction

Confidence in model computations of environmental systems depends on:

- (1) **The quality of the models:** Real-life process-to-model translations may vary between disciplines and scientists. In addition errors in the model structure, model parameter estimation and numerical computation methods are a few of the causes that affect confidence in model predictions. One of the causes of uncertainty in prior assumptions about a system is directly related to the problem of aggregation, i.e., to what level of detail should a system's process be modeled. Processes may be described in a process-oriented way or reflected by empirical relationships. All models presented in the Mapping Manual and Sverdrup *et al.* (1990) may be considered process oriented on the whole, although some of them have empirical characteristics. Empirical (sub)models consist of a simple relationship between a (sub)process' output and input, treating the intermediate processes as a black box.

However there exists no common opinion on the degree of detail by which acidifying processes should be described. As a result, different models exist which aim at describing similar processes (e.g., MAGIC, ILWAS, SMART). The distinction made above, between process-oriented (so-called 'mechanistic' or 'complex') models and less process-oriented, more empirical models (also called 'statistical' or 'response-surface' models) may postpone scientific agreement on methods to represent environmental systems in general and critical loads in particular. Concerning water quality modeling, Beck (1987, p. 1396) argues that "...detailed spatial patterns of water circulation and equally detailed differentiation of ecological behavior described by the more complex models would demand experimental observations that are simply not technically feasible." This statement also applies to critical load models. Different viewpoints about the critical load models to apply imply that it will be difficult to agree on one single method (and data requirements) to be used throughout Europe. The consequence is that results will have to be interpreted including a range of uncertainty, which results from the application of different models (and data), rather than discussing which model performs best.

- (2) **The quality of the data:** Data should be sampled such that ecosystem processes can properly be monitored and reflected. In turn, models which use the sampled data, should be designed to reflect the properties of the system. The wishes to (1) monitor an ecosystem and (2) obtain data for a model representing the ecosystem are difficult to be translated into a sampling strategy. It is one thing to design a sampling strategy which represents the real process and another to have the data suit the model design which is believed to describe the processes involved. The conflicts between models and data lead to uncertainties as a result of e.g., (1) the number of sampled ecosystem processes is not large enough to represent the ecosystem, (2) the variables sampled are not representative of the processes, (3) the model designed to represent the system may be too detailed and require more data or (4) the model is not detailed enough to treat all the available data. The amount of sites to monitor, the stratification of the sites, and the level of detail of the system parameters to be sampled, may all contribute to potential errors in understanding a system.

The quality of the data needed for the computation of critical loads varies over ecosystems and over countries. The weathering rate, for example, is a variable which is very important for the computation of critical loads but for which the required measuring methods (i.e., total (soil) analysis) have not been sampled to cover Europe. Empirical methods (see Appendices 1 and 3) are used instead. Another source of uncertainty in the assessment of critical loads is due to the application of different critical chemical values which may lead to errors in the computation of

critical loads for a given receptor.¹ An illustration of the consequence of the uncertainty in critical chemical values can be found in de Vries (1991) and Sverdrup *et al.* (1990). Suggested averages of critical chemical values are presented in Table 4.1 (see also Table 1.2 in Sverdrup *et al.* 1990). **It is recommended to use these values.**

In summary, uncertainties due to a varying quality of data are partly a consequence of a natural range in sensitivities of indicator organisms and partly reflects the lack of knowledge regarding the effect of acid deposition.

- (3) **The extrapolation of the results:** The uncertainties mentioned above may combine for unexpected results. This is especially true when results apply to broad spatial scales (e.g., Europe) or long temporal scales (e.g., time needed to reach critical chemical values when critical loads are exceeded).

It is clear from the methods described in previous chapters that the accuracy of maps of critical loads and levels may suffer from each of the components of uncertainty mentioned above. A way to deal with uncertainty is to introduce a probabilistic component in modeling (see also Mihram, 1972, p. 12; Hettelingh, 1990; pp.13, 49-55; Hettelingh *et al.*, 1992).

In what follows, this chapter will focus and summarize the notion and application of uncertainty to two issues encountered while producing maps of critical loads and levels, i.e., the treatment of uncertainty about (1) models and model parameters, and to (2) spatial and temporal scales.

5.2 Uncertainty

5.2.1 Uncertainty in modeling and model parameters

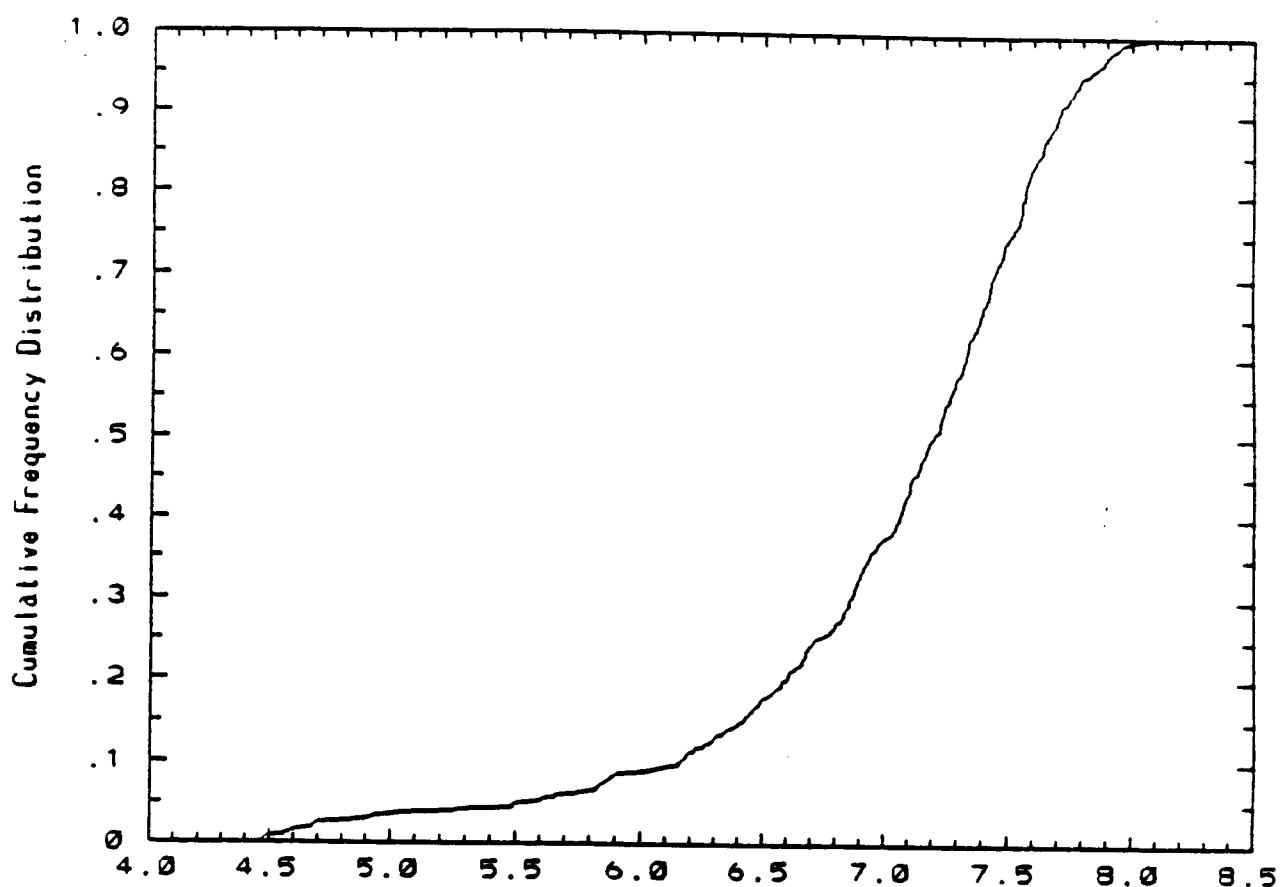
When applying uncertainty analysis to a model, the influence of the variance of model parameters (i.e., right-hand side variables in equations) on the model response is investigated. Thus, uncertainty in model response is expressed as the variance of the model response. The investigation of model uncertainty is often confused with sensitivity analysis of a model. The distinction between uncertainty and sensitivity analysis is that the latter describes the effect of an arbitrarily small distortion in a parameter on the change of the model response.

The contribution of individual parameter errors to the uncertainty in model results may numerically be investigated by the Monte Carlo method². This method implies the use of a random generator for sampling parameter values from a predefined range in order to solve the model.

In practice this means that a large number, e.g., 500, samples of independent parameter values are consecutively used as input to the model, leading to 500 model responses. From this set of model responses the frequency of occurrences can be derived. The application of the Monte Carlo method may be applied for both steady state and dynamic models. In the latter case, for each sample that is drawn in a starting year, the model continues to simulate the process through time.

An example of the cumulative frequency distribution, including the statistics, of the pH response from 500 runs with the RAINS Lake Module is shown in Figure 5.1. The figure shows that the probability of a pH lower than 6.0, for example, occurs in about 10% of the runs and that in 50% (median) of the samples the pH will lie below 7.2 (7.198 in Figure 5.1).

-
1. For example, the range of aluminum toxicity appears to be very large for different tree species.
 2. An overview of the literature on uncertainty analysis and on the theory and application of Monte Carlo methods can be found in Hettelingh, 1989 (pp. 49-68).



Mean	:	7.019
Absolute deviation	:	0.5288
Standard deviation	:	0.7125
Variance	:	0.5077
Skewness	:	-1.523
Kurtosis	:	2.514
Minimum	:	4.464
2.5% percentile	:	4.703
25% percentile	:	6.712
Median	:	7.198
75% percentile	:	7.502
97.5% percentile	:	7.896
Maximum	:	8.070
Number of iterations	:	500

Figure 5.1. Example of a cumulative distribution of lake pH based on 500 Monte Carlo runs with the RAINS lake module (from Hettelingh, 1990, p. 217).

The influence of a particular model parameter on the variance of the model response may vary among all the model parameters. Some parameters explain more of the response variance than others. Thus, the set of model parameters can be hierarchically ordered from very important (explaining most of the response variance) to irrelevant (explaining very little or nothing of the response variance). This distinction between the importance of model parameters becomes particularly interesting when compared to the quality of the data. If for example, the weathering explains 75% of the variance in pH, but the data on weathering are not complete or subject to doubt, particular care is needed in the interpretation of the pH results. In such a case it is advisable to run the model a number of times for a number of weathering values, which apply to the particular site or grid square, to obtain a range of pH predictions.

Since different models may have a varying hierarchy of important parameters, not all the models are equally suited to be applied on all the European ecosystems. Comparing models to one another can be done by Monte Carlo analysis as has been shown in Rose *et al.* (1990).

5.2.2 Spatial generalization to regions

Currently, critical loads are computed and displayed in grid squares as the chosen unit for mapping. However, the use of grid squares rather than polygons (regions) introduces uncertainty in the interpretation of displayed critical load. The display of the 5- or 50-percentile critical load (see Section 2.5) in a $1^\circ \times 0.5^\circ$ grid may look different compared to the same display in the EMEP grid. The reason is that the probability of the occurrence of broad ranges of critical loads increases as the grid square covers more surface. Therefore, large grid squares have an increased probability of high critical loads in 5% or 50% of the area within the grid square. Small grid squares will generally cover a small range of critical loads.

The conclusion is that the application of rastered maps leads to loss of information, especially when the raster consists of large grid squares.

In the mapping exercise, effort should be put in improving the spatial display of ecosystem sensitivity and thus reduce uncertainty introduced by rastering (see above). Data is already available for some ecosystems in Europe in the form of spatially scattered samples of related ecosystems (e.g., a region containing lakes or a forested region). The use of related data points will improve the geographical interpretation of critical loads. Application of gravitation techniques within Geographical Information Systems will allow for the production of polygons (regions) which are less arbitrary than grid squares.

Models have often been developed for particular sites and the extrapolation to larger areas introduces an additional source of error to the uncertainties mentioned above. Recently two volumes (Kämäri (ed.), 1990; Kämäri *et al.* (eds.), 1990) have been published containing research results on the regional application of acidification models. The procedure of regionalization generally consists of (a) a calibration of the model to a regional distribution of model response (b) the application of the calibration result to obtain model predictions for different time periods (see also Hettelingh, 1990; Posch and Kämäri, 1990; Sutton, 1990; Gardner *et al.*, 1990).

The application of calibration procedures, i.e., finding a set of model parameters that lead to a desired model output, may introduce inaccuracies (Sutton, 1990). If, additionally, a calibration result obtained in a particular region is applied in a different region, errors may occur due to the heterogeneity of the regional ecosystems (see Gardner *et al.*, 1990; Hettelingh, 1990, pp. 94-133; Hettelingh *et al.*, 1990).

It is recommended to ensure **homogeneity** of the regions to which models are applied (e.g., similar ecosystems, similar soil types). The maps of critical loads would in the latter case consist of a few shaded regions, each provided with a cumulative distribution of the model results obtained with the Monte Carlo method.

Homogeneity may also be ensured by inspecting the relationship between the model output and the most important model parameters (see Hettelingh, 1990, pp. 134-169) in an arbitrarily bounded region (e.g., a country). The method leads to a varying subdivision of an arbitrarily bounded region into subregions (**flexible zones**) which are homogeneous with respect to the most important model parameters, and acid deposition.

The use of dynamic models (SMART, SAFE) in combination with techniques mentioned above will lead to results which apply to homogeneous regions (i.e., with only one type of ecosystem) rather than to heterogeneous grid squares.

5.2.3 Temporal assessments

Critical loads, according to definition, are steady in time. However, maps of critical loads will be used for the assessment of abatement strategies, by comparing the excess of critical loads both now and in the future. Exceedances will occur in some parts of Europe, in spite of emission abatements reached. It is necessary to anticipate ways to deal with this problem, even though it is not directly related to our current involvement, i.e., the production of critical load maps.

Critical loads will not change in time, but chemical values will change as a result of deposition changes in the future. A large current excess of critical loads may lead to chemical values becoming critical not before some time in the future. On the other hand, a small exceedance may already lead to unacceptable chemical values in the near future. Such results may be computed by using one of the dynamic models suggested in the Mapping Manual (see Hettelingh, 1990, pp. 134-169; Hettelingh *et al.*, 1992).

5.2.4 Concluding remarks

1. Large grid squares have an increased probability of high critical loads in 5% or 50% of the area within the grid square. Small grid squares will generally cover a small range of critical loads. The result is that an x-percentile critical load in large grid squares will tend to exceed the x-percentile critical load in smaller grid squares. Therefore, the application of rastered maps leads to loss of information, especially when the raster consists of large grid squares.
2. Uncertainty analysis of steady state as well as dynamic models will provide insight in the confidence of model results for different ecosystems in a grid square. The Monte Carlo method is a convenient tool.
3. Using uncertainty analysis the most important model parameter (i.e., explaining most of the variance of model output) will be distinguished. If the data input for the most important variable is dubious, the presentation of deterministic model solutions should be avoided. Instead a cumulative distribution of model response (critical loads) in a grid square should be provided as the result of the repeated input out of a range of values for the most important, or all, variables. The map will consist of a shaded grid squares. The shading class is chosen according to the percentile that is displayed.
4. If information is lacking to provide model predictions in every single grid square, cumulative distributions of model responses in a bounded region should be provided. An effort should be made to divide such a region into homogenous subregions (soil types, ecosystems, flexible zones).
5. If the distinction of homogeneous subregions is too cumbersome, an effort should be made to make a classification of the occurrence of sensitive areas within a bounded region. The

distinction of sensitivity classes may vary for different models, which leads to the recommendation to restrict to a physical classification criterion.

If the latter classification is difficult to obtain, then the model should be made subject to Monte Carlo simulation by sampling from the data ranges of each of the model parameters. The result will be a cumulative distribution as presented in Figure 5.1 in which model output cannot be attributed to a particular ecotype within the region. The map will consist of a set of shaded bounded regions. Shading patterns will be chosen according to the chosen percentile of model outputs. For each shaded region the cumulative distribution is provided displaying the frequency of each sensitivity class (i.e., a particular range of model results).

6. If no uncertainty analysis is applied, and not all data values are known exactly, the model should at least be run for the upper and lower limits of the data range(s) in question. In this case cumulative distributions should reflect the percentage of the area in which a particular model response range is obtained.
7. Predictions in a target year of (critical) chemical values could also be represented in a cumulative frequency distribution. This future cumulative distribution is obtained by repeatedly running the model from a base year to the target year. For every run, another set of parameter input values is used. Each set of input parameter values is obtained from the Monte Carlo samples that were obtained in the base year. The model run is completed once the predictions for the target year are obtained. Thus, a great number (e.g., 500) predictions for the target year may be displayed in a cumulative distribution.
8. Finally it should be noted that due to an existing collaboration between the RIVM and the Oak Ridge National Laboratory (USA), methods and software developed by Gardner *et al.* (1983) for the application of Monte Carlo sampling are available at the Coordination Center. National Focal Centers are invited to use the methods or have them applied by the Coordination Center.

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APPENDIX 1. THE 'LEVEL 0' APPROACH FOR CALCULATING CRITICAL LOADS ¹

'Level 0' approaches are semi-quantitative methods using existing geographical databases to assess the sensitivity of ecosystems to acidic deposition to which critical loads values may be applied. An example of a semi-quantitative assessment is the map of relative sensitivity to acidic deposition, which has been produced at the Stockholm Environmental Institute (Kuylenstierna and Chadwick, 1990), to which target loads have been applied on the basis of critical loads information in the literature. This 'Level 0' approach uses the information given in the Skokloster critical loads document (Nilsson and Grennfelt, 1988).

In Nilsson and Grennfelt (1988) soil types are assigned to one of five classes according to mineralogy of parent rock-type (Table A1.1). To each class a critical load is suggested (Table A1.2). Table A1.3 describes how other factors such as land use, amount of rainfall etc., may modify critical load values as specified in Table A1.2. When a combination of factors (Table A1.3) enhances sensitivity of the soil, the lower limit of the critical load range from Table A1.2 should be used. Otherwise (for examples, under conditions which mitigate soil sensitivity), the upper limit of the ranges should be applied. This approach has been used in the United Kingdom.

Table A1.1. Mineralogy and petrological classification of soil material.

Class	Minerals controlling weathering	Usual parent rock
1	Quartz K-feldspar	Granite Quartzite
2	Muscovite Plagioclase Biotite (< 5%)	Granite Gneiss
3	Biotite Amphibole (< 5%)	Granodiorite Greywakee Schist Gabbro
4	Pyroxene Epidote Olivine (< 5%)	Gabbro Basalt
5	Carbonates	Limestone Marlstone

Source: Nilsson and Grennfelt (1988), p. 13.

1. Written in collaboration with Johan Kuylenstierna from the University of York.

Table A1.2. Critical load for forest soils (0-50 cm).

Class	Total acidity (kmol H ⁺ km ⁻² yr ⁻¹)	Equivalent amount of sulphur (kg ha ⁻¹ yr ⁻¹)
1	< 20	< 3
2	20-50	3-8
3	50-100	8-16
4	100-200	16-32
5	> 200	> 32

Source: Nilsson and Grennfelt (1988), p. 14.

Table A1.3. Conditions influencing critical loads to forest soils.

Factor	Decreasing critical load value	Increasing critical load value
Precipitation	high	low
Vegetation	coniferous	deciduous
Elevation/slope	high	low
Soil texture	coarse-sandy	fine
Soil/till depth	shallow	thick
Soil sulphate	low	high
adsorption capacity		
Base cation deposition	low	high

Source: Nilsson and Grennfelt (1988), p. 14.

Thus in the United Kingdom national soils data have been assigned to five classes according to the Skokloster report, although certain modifications have been made, especially concerning areas of peat. It is intended to combine land use information with the soil data to modify the critical loads derived. The critical loads derived in this way compare favorably with loads calculated using the Henriksen steady state water chemistry method for the same areas (see also Hettelingh *et al.*, 1991).

Therefore, an approach for a country could be:

1. Assign national soil data (mapped or in data base) to one of five 'Skokloster soil classes' of Table A1.1;
2. Modify the classification by considering the factors suggested in Table A1.3;
3. Assign critical loads given in Table A1.2.

The Skokloster document does not define a procedure for modification using additional factors. Therefore, a certain amount of interpretation may be necessary in the creation of the 'Level 0' critical loads map. The method could be refined by consulting with the Stockholm Environmental Institute. The classification may need to be modified to take into account local conditions. For example, soils in southern Europe having high sulphate adsorption capacities would lead to a modified classification of soils into weathering rate classes.

APPENDIX 2. CRITICAL LOADS OF ACIDITY, SULPHUR AND NITROGEN AND THE COMPUTATION OF CRITICAL LOAD EXCEEDANCES

This appendix summarizes the equations used to compute critical loads, present loads, and the excess of the critical loads of acidity, sulphur and nitrogen. The 'critical load' for sulphur and nitrogen will be discussed using a meteorological approach and an ecosystems approach. The relationship between the formulation of a critical load for sulphur and for nitrogen is discussed in relation to the eutrophying and acidifying aspects of nitrogen. An effort is thus made to harmonize critical loads for sulphur and nitrogen (nutrient and acidity) with the critical load for actual acidity.

1. Critical Loads of Acidity

The following formulations of critical loads for acidity can be distinguished (see also section 4.2.2):

- *Critical load of sulphur and nitrogen:*

$$CL(S+N) = BC_{td}^* + BC_w - BC_u + N_u + N_{i(acc)} - ANC_{l(acc)} \quad (2.1)$$

where:

CL(S+N) = Critical load of sulphur and nitrogen
 BC_{td}^* = Seasalt-corrected total base cation deposition
 BC_w = Base cation weathering
 BC_u = Base cation uptake
 N_u = Nitrogen uptake
 $N_{i(acc)}$ = Acceptable nitrogen immobilization
 $ANC_{l(acc)}$ = Acceptable alkalinity leaching

- *Critical load of potential acidity:*

$$CL(AC_{pot}) = BC_w - BC_u + N_u + N_{i(acc)} - ANC_{l(acc)} \quad (2.2)$$

where:

$CL(AC_{pot})$ = Critical load of potential acidity ¹

- *Critical load of actual acidity:*

$$CL(AC_{act}) = BC_w - ANC_{l(acc)} \quad (2.3)$$

where:

$CL(AC_{act})$ = Critical load of actual acidity

- *Critical load of acidity:*

1. NH_4 is included as a potential acid since NH_4 produces two protons in the soil through nitrification.

Anticipating the formulation of a critical load for sulphur and nitrogen in section 3 of this appendix, the following formulation describes the critical load of acidity:

$$CL(Ac) = CL(Ac_{act}) + N_u + N_{i(acc)} \quad (2.4)$$

or:

$$CL(Ac) = BC_w + N_u + N_{i(acc)} - ANC_{l(acc)} \quad (2.5)$$

where:

$CL(Ac)$ = Critical load for acidity

2. Present Loads of Acidity

According to the critical load equations, present load equations are defined as:

- *Present load of sulphur and nitrogen:*

$$PL(S+N) = PL(SO_x) + PL(NO_x) + PL(NH_x) \quad (2.6)$$

where:

$PL(S+N)$ = Present load of sulphur and nitrogen

$PL(SO_x)$ = Present load of sulphur

$PL(NO_x)$ = Present load of nitrogen oxides

$PL(NH_x)$ = Present load of ammonia and ammonium

- *Present load of potential acidity:*

$$PL(Ac_{pot}) = PL(S+N) - BC_{dt}^* \quad (2.7)$$

where:

$PL(Ac_{pot})$ = Present load of potential acidity

- *Present load of actual acidity:*

$$PL(Ac_{act}) = PL(Ac_{pot}) + BC_u - N_u - N_{i(acc)} \quad (2.8)$$

where:

$PL(Ac_{act})$ = Present load of actual acidity

- *Present load of acidity:*

Anticipating the formulation of the critical load for sulphur and nitrogen (see section 3 of this appendix) and taking into account the necessity to compute exceedances from the critical load of acidity (see equation 2.5), the following present load of acidity is introduced:

$$PL(Ac) = PL(Ac_{act}) + N_u + N_{l(acc)} \quad (2.9)$$

3. Critical Load for Sulphur and Nitrogen

The WGS, WGE, TFM and TFIAM have repeatedly stressed the necessity of computing a critical load for sulphur to be used in the formulation of a new sulphur protocol. Until now the term 'impact load' was used to stress the fact that no definition could be provided for a critical load of sulphur which would be consistent with the critical load for acidity. However, to ease communication, the term 'critical load for sulphur' will be used in this appendix to describe a variable which is computed as a portion of the critical load of acidity generated by sulphur. During the first two CCE training sessions, two methods were discussed: (1) the ecosystem-based method and (2) the deposition-based method.

It was decided that the deposition-based method should first be applied in all countries. Both methods are discussed in the following.

3.1 The ecosystem-based method

In this approach, an ecosystem-based critical load for sulphur, $CL(S_e)$, is derived from the critical load of potential acidity (which includes sulphur and nitrogen) according to de Vries (1991):

$$CL(S_e) = CL(S + N) - CL(N_e) \quad (2.10)$$

Where $CL(N_e)$ is the ecosystem-based critical load for nitrogen with respect to eutrophication, defined in Sverdrup *et al.* (1990) and de Vries (1991) as:

$$CL(N_e) = N_u + N_{l(acc)} + N_{l(acc)} \quad (2.11)$$

where:

$N_{l(acc)}$ = Acceptable nitrogen leaching. (In section 3.2 a point will be made to consider the acceptable nitrogen leaching in relation to the acidification caused by nitrogen.)

Substitution of equations 2.2 and 2.11 into equation 2.10 gives:

$$CL(S_e) = BC_{td}^* + BC_w - BC_u - (ANC_{l(acc)} + N_{l(acc)}) \quad (2.12)$$

3.2 The deposition-based method

3.2.1 Sulphur

In this approach the critical load of the acidity input by sulphur is related to the critical load of actual acidity (equation 2.3) as follows:

$$CL(S_d) = S_f \cdot CL(Ac_{act}) \quad (2.13)$$

where:

$CL(S_d)$ = Critical load of sulphur (deposition-based method)
 S_f = Sulphur fraction

In the report of the second CCE training session, a sulphur fraction was defined as the ratio between sulphur deposition and the summed depositions of sulphur and nitrogen. The report also mentions that the sulphur fraction should be equal to one when the nitrogen deposition is smaller than the sum of nitrogen uptake and nitrogen immobilization. However, there is no good reason why the correction for immobilization and uptake should not be introduced when nitrogen deposition exceeds uptake and immobilization. Consequently, the formulation of the sulphur fraction becomes:

$$S_f = \begin{cases} \frac{PL(SO_x)}{PL(SO_x) + PL(NO_x) + PL(NH_x) - N_u - N_{i(crit)}} & \text{if } PL(NO_x) + PL(NH_x) \geq N_u + N_{i(crit)} \\ 1 & \text{otherwise} \end{cases} \quad (2.14)$$

3.2.2 Nitrogen

Equation 2.11 provides a definition of the critical load for nitrogen as nutrient in which the acceptable nitrogen leaching rate is related to eutrophication. However, it is also possible to derive a critical load for nitrogen which is associated to acidity. This is obtained by assuming that the excess of nitrogen deposition to uptake and immobilization will cause acidification. A deposition-based critical load for nitrogen, $CL(N_d)$, i.e., the acidity input by nitrogen, becomes:

$$CL(N_d) = N_u + N_{i(acc)} + (1 - S_f) \cdot CL(Ac_{act}) \quad (2.15)$$

By defining a deposition-based critical load of acidity, $CL(Ac)$, according to:

$$CL(Ac) = CL(S_d) + CL(N_d) \quad (2.16)$$

and by substitution of equations 2.13 and 2.15 in equation 2.16, we obtain (cf. equation 2.4):

$$CL(Ac_{act}) = CL(Ac) + N_u + N_{i(acc)} \quad (2.17)$$

4. Present Loads of Sulphur and Nitrogen

4.1 The ecosystem-based method

In order to be able to compute the excess of the critical load of acidity associated with sulphur in the ecosystem-based method, the present load of sulphur is defined as:

$$PL(S_e) = PL(S + N) - PL(N_e) \quad (2.18)$$

where:

$PL(S_e)$ = Present load of sulphur (ecosystem-based method)

$PL(N_e)$ = Present load of nitrogen (ecosystem-based method), which is defined as:

$$PL(N_e) = PL(NO_x) + PL(NH_x) \quad (2.19)$$

Substitution of equations 2.6 and 2.19 into equation 2.18 leads to:

$$PL(S_e) = PL(SO_x) \quad (2.20)$$

4.2 The deposition-based method

In order to be able to compute an excess of the critical load of acidity associated with sulphur in the deposition-based method, a decision has to be made about the portion of the net acidity input by base cation uptake and base cation deposition to be attributed to sulphur. This is done by multiplying the base cation deposition and base cation uptake by the sulphur fraction:

$$PL(S_d) = PL(SO_x) + S_f \cdot (BC_u - BC_d) \quad (2.21)$$

where:

$PL(S_d)$ = Present load of sulphur (deposition-based method)

The remaining part can be attributed to acidity associated with nitrogen:

$$PL(N_d) = PL(NO_x) + PL(NH_x) + (1-S_f) \cdot (BC_u - BC_d) \quad (2.22)$$

where:

$PL(N_d)$ = present load of nitrogen (deposition-based method)

By substituting equations 2.21 and 2.22 into equations 2.6 through 2.8, the following formulation of the present load of actual acidity is obtained¹:

$$PL(Ac_{act}) = PL(S_d) + PL(N_d) - N_{l(acc)} - N_u \quad (2.23)$$

By defining a present load of acidity, $PL(Ac)$, according to:

$$PL(Ac) = PL(S_d) + PL(N_d) \quad (2.24)$$

and substituting equation 2.24 into equation 2.23, we obtain (cf. equation 2.9):

$$PL(Ac_{act}) = PL(Ac) - N_{l(acc)} - N_u \quad (2.25)$$

5. The Excess of Critical Loads for Acidity

The excess of critical load, $CL_{(exc)}$, is defined as:

$$CL_{(exc)} = PL - CL \quad (2.26)$$

Irrespective of the use of the various formulations for acidity, i.e., sulphur plus nitrogen, potential acidity, actual acidity, or acidity, this excess is calculated as:

$$CL(Ac)_{exc} = PL(SO_x) + PL(NO_x) + PL(NH_x) - BC_{dt}^* - BC_w + BC_u - N_u - N_{l(acc)} + ANC_{l(acc)} \quad (2.27)$$

The definitions that are used specifically are the critical load of actual acidity and the critical load of acidity (N + S using the deposition-based method). It can be proven that the excess in these acid loads are equal, i.e.:

$$CL(Ac_{act})_{exc} = PL(Ac_{act}) - CL(Ac_{act}) \quad (2.28)$$

1. Also note that the sulfur fraction can be expressed as $S_f = PL(S) / PL(Ac_{act})$.

Substitution of equations 2.23 and 2.17 into equation 2.28 leads to:

$$CL(Ac_{act})_{exc} = PL(S_d) + PL(N_d) - N_{i(acc)} - N_u - (CL(Ac) - N_u - N_{i(acc)}) \quad (2.29)$$

which is equal to:

$$CL(Ac_{act})_{exc} = PL(S_d) + PL(N_d) - CL(Ac) \quad (2.30)$$

Substitution of equation 2.9 into equation 2.30 leads to:

$$CL(Ac_{act})_{exc} = PL(Ac) - CL(Ac) \quad (2.31)$$

6. The Excess of Critical Loads of Sulphur and Nitrogen

6.1 The ecosystem-based method

The excess of the critical load for sulphur, $CL(S_e)_{exc}$, is defined as:

$$CL(S_e)_{exc} = PL(S_e) - CL(S_e) \quad (2.32)$$

Substitution of equations 2.12 and 2.20 into equation 2.32 yields:

$$CL(S_e)_{exc} = PL(SO_x) - BC_{dt}^* - BC_w + BC_u + ANC_{l(acc)} + N_{l(acc)} \quad (2.33)$$

The excess of the critical load for nitrogen, $CL(N_e)_{exc}$, is defined as:

$$CL(N_e)_{exc} = PL(N_e) - CL(N_e) \quad (2.34)$$

Substitution of equations 2.11 and 2.19 into equation 2.34 yields:

$$CL(N_e)_{exc} = PL(NO_x) + PL(NH_x) - N_u - N_{i(acc)} - N_{l(acc)} \quad (2.35)$$

Contrary to the deposition-based method, a critical load exceedance using the ecosystem-based method is calculated by directly comparing the present deposition levels of sulphur (cf. equation 2.20) and nitrogen (equation 2.19) with the critical deposition levels.

Substitution of equations 2.33 and 2.35 in equation 2.27 gives:

$$CL(Ac)_{exc} = CL(S_e)_{exc} + CL(N_e)_{exc} \quad (2.36)$$

i.e., the sum of the excess in critical loads for sulphur and nitrogen equals the excess of the critical acid load.

6.2 The deposition-based method

For the derivation of critical loads of sulphur and its excess, the deposition-based method has presently been used (Hettelingh *et al.* 1991). Using this approach the critical load excess for sulphur can be derived by substituting equations 2.13 and 2.21 into equation 2.32:

$$CL(S_d)_{exc} = PL(SO_x) + S_f \cdot (BC_u - BC_d) - S_f \cdot CL(Ac_{act}) \quad (2.37)$$

which, after rearranging leads to:

$$CL(S_d)_{exc} = PL(SO_x) + S_f \cdot (BC_u - BC_d - CL(Ac_{act})) \quad (2.38)$$

Using the deposition-based method, the excess of the critical load for nitrogen can be derived by substitution of equations 2.15 and 2.22 into equation 2.34:

$$CL(N_d)_{exc} = PL(NO_x) + PL(NH_x) + (1-S_f) \cdot (BC_u - BC_d) - N_u - N_{i(acc)} - (1-S_f) \cdot CL(Ac_{act}) \quad (2.39)$$

which, after rearranging leads to:

$$CL(N_d)_{exc} = PL(NO_x) + PL(NH_x) + (1-S_f) \cdot (BC_u - BC_d - CL(Ac_{act})) - N_u - N_{i(acc)} \quad (2.40)$$

The treatment of separate critical loads of sulphur and nitrogen is consistent with the critical load of actual acidity (cf. equation 2.31) since:

$$CL(Ac_{act})_{exc} = PL(Ac) - CL(Ac) \quad (2.41)$$

which after rearrangement leads to:

$$CL(Ac_{act})_{exc} = (PL(S_d) - CL(S_d)) + (PL(N_d) - CL(N_d)) \quad (2.42)$$

Equation 2.42 indicates that the excess of the critical load of actual acidity is equal to the sum of the excess of the critical load of sulphur and the excess of the critical load of nitrogen. This can be seen by substitution of equations 2.32 and 2.34 into equation 2.42 leading to:

$$CL(Ac_{act})_{exc} = CL(S_d)_{exc} + CL(N_d)_{exc} \quad (2.43)$$

APPENDIX 3. PROPOSAL TO ESTIMATE WEATHERING RATES FROM SOIL TYPE INFORMATION ON THE FAO-UNESCO SOIL MAP OF EUROPE

At the first CCE Training Session some discussion emerged from the question how to estimate weathering rates when sufficient field data are lacking to have the weathering rate computed (i.e., by means of PROFILE). This appendix proposes a method for estimating weathering rates based on available soil type information.

3.1 Principles

The code of a mapping unit on the FAO soil map consists of four types of information:

- | | | |
|-----------------------------|-------------------------|----------|
| - the dominant soil unit | (1- or 2-letter symbol) | e.g., Po |
| - the associated soil units | (1- or 2-digit symbol) | e.g., 25 |
| - the texture class | (1-digit symbol) | 1, 2, 3 |
| - the slope class | (1-letter symbol) | a, b, c |

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

In this proposal, 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 to differentiate among mapping units. Furthermore, the slope class is considered irrelevant regarding the weathering rate.

The dominant parent material of each soil type is converted into three different classes; i.e., (1) acidic, (2) intermediate, and (3) basic, with the following relationships:

- (1) Acidic: Sand(stone), gravel, granite, quartzite, gneiss (schist, shale, greywacke, glacial till);
(2) Intermediate: Granodiorite, loess, fluvial and marine sediments (schist, shale, greywacke, glacial till);
and
(3) Basic: Gabbro, basalt, dolomite, volcanic deposits.

Schist, shale, greywacke, and glacial till are put between brackets both in (1) and (2). A soil type containing these parent materials can be converted to both classes, depending on the other parent materials available.

Apart from these three classes given above, two additional classes have been defined where silicate weathering is taken as zero; i.e.:

- (0) Peat soils: These soils do not contain silicates nor Al-hydroxides and the pH in these soils is completely determined by H^+ /base cation exchange;
(4) Calcareous: The pH in these soils is completely determined by soils: carbonate weathering (equilibrium).

When determining critical acid loads, the carbonate weathering rate of calcareous soils should be given. However, since this weathering rate always equals the acid load, it might be better to exclude calcareous soils and just give the percentage of forests on these soils.

The assumed dominant parent material for each soil type on the FAO soil map of Europe below forests (81) is given in Table A3.1. Classes 0 (peat) and 4 (calcareous soils, marl, limestone) are also added. The conversion is based on the lithology given in Table A3.3 of Volume V of the FAO soil map of the world (Europe). An explanation of the various codes is given in Table A3.2.

Table A3.1. Proposed conversion between soil type and parent material class.

0	1	2	3	4
Od	Ao	Bv		Bc
Oe	Bd	Ch	Th	Be
	Be	Cl	Tm	Bg
	Bh	Gd	To	Bk
	Dd	Ge	Tv	Ck
	De	Gh		E
	Dg	Gm		Hc
	I			Hg
	Pg	Hh		Jc
	Ph	Hi		Kk
	Pl	Je		Lc
	Po	Kh		Lv
	Pp	Kl		Rc (1)
	Qc	Lf		So
	Ql	Lg		Sm
	Re	Lo		Vc
	U	Mo		Xk
	Wd	Vp		Xy
	I-Bd	We		Zg
	I-Be			Bc-Lc
	I-L			I-Bc
	I-Be-Lc			I-Bc-Lc
	I-Bc-U			I-C
	I-Lo-Bc			I-E
	I-Po			I-Lc
	I-Po-Od			I-Lc-E
	I-Re-Rx			I-Re-Xk
	I-U			
	Lo-Lc			

1. Rc is differentiated into Rca, Rcb, and Rcc.

Table A3.2. Soil units for Europe.

J FLUVISOLS	T ANDOSOLS	C CHERNOZEMS	D PODZOLUVISOLS
Je Eutric Fluvisols	To Ochric Andosols	Ch Haplic Chernozems	De Eutric Podzoluvisols
Jc Calcaric Fluvisols	Tm Mollic Andosols	Ck Calcic Chernozems	Dd Dystric Podzoluvisols
Jd Dystric Fluvisols	Th Humic Andosols	Cl Luvic Chernozems	Dg Gleyic Podzoluvisols
Jt Thionic Fluvisols	Tv Vitric Andosols	Cg Glossic Chernozems	
G GLEYSOLS	V VERTISOLS	H PHAEZEMS	P PODZOLS
Ge Eutric Gleysols	Vp Pellic Vertisols	Hh Haplic Phaeozems	Po Orthic Podzols
Gc Calcaric Gleysols	Vc Chromic Vertisols	Hc Calcaric Phaeozems	Pl Leptic Podzols
Gd Dystric Gleysols		Hi Luvic Phaeozems	Ph Humic Podzols
Gm Mollic Gleysols	Z SOLONCHAKS	Hg Gleyic Phaeozems	Pp Placic Podzols
Gh Humic Gleysols	Zo Orthic Solonchaks		Pg Gleyic Podzols
Gx Gelic Gleysols	Zm Mollic Solonchaks	M GREYZEMS	W PLANOSOLS
	Zg Gleyic Solonchaks	Mo Orthic Greyzems	We Eutric Planosols
R REGOSOLS	S SOLONETZ	B CAMBISOLS	Wd Dystric Planosols
Re Eutric Regosols	So Orthic Solonetz	Be Eutric Cambisols	
Rc Calcaric Regosols	Sm Mollic Solonetz	Bd Dystric Cambisols	A ACRISOLS
Rd Dystric Regosols	Sg Gleyic Solonetz	Bh Humic Cambisols	Ao Orthic Acrisols
Gx Gelic Regosols		Bg Gleyic Cambisols	Af Ferric Acrisols
	Y YERMOSOLS	Bk Calcic Cambisols	Ah Humic Acrisols
I LITHOSOLS	Yk Calcic Yermosols	Bc Chromic Cambisols	
	X XEROSOLS	Bv Vertic Cambisols	O HISTOSOLS
Q ARENOSOLS	Xh Haplic Xerosols	L LUVISOLS	Oe Eutric Histosols
Qc Cambic Arenosols	Xk Calcic Xerosols	Lo Orthic Luvisols	Od Dystric Histosols
Ql Luvic Arenosols	Xy Gypsic Xerosols	Lc Chromic Luvisols	Ox Gelic Histosols
	Xl Luvic Xerosols	Lk Calcic Luvisols	
E RENDZINAS		Lv Vertic Luvisols	
U RANKERS	K KASTANOZEMS	Lf Ferric Luvisols	
	Kh Haplic Kastanozems	La Albic Luvisols	
	Kk Calcic Kastanozems	Lg Gleyic Luvisols	
	Kl Luvic Kastanozems		

Information on the various texture classes on the map is given in Table A3.3.

Table A3.3. Texture classes on the FAO soil map of Europe.

Code	Description	Clay content (%)	
		range	average
1	coarse (sandy)	< 18	8
2	medium (loamy)	18-35	25
3	fine (clayey)	> 35	50
1/2	coarse to medium	< 35	15
1/3	coarse to fine *	n/a	25
2/3	medium to fine	> 18	40

3.2 Proposal

For 1-meter depth and for a root zone of the proposed weathering rate classes in $\text{mol}_e \text{ ha}^{-1} \text{ yr}^{-1}$ are given in Table A3.4. The various classes are based on the Skokloster workshop report on "Critical Loads of sulphur and nitrogen" (Nilsson and Grennfelt, 1988).

Table A3.4. Proposed weathering rates ($\text{mol}_e \text{ ha}^{-1} \text{ yr}^{-1} \text{ m}^{-1}$).

Weathering Rate Class	Weathering Rate (depth)	Root Zone asm
1	500	250
2	1000	500
3	1500	750
4	2000	1000
5	2500	1250
6	3000	1500

The proposed conversion table between parent material class, texture class and weathering rate class is given in Table A3.5.

Table A3.5. Proposed weathering rate classes for various combinations of parent material class and texture class.

Parent Material Class	Texture class					
	1	1/2	1/3	2	2/3	3
1	1	2	–	3	4	–
2	2	3	4	4	5	6
3	2	3	–	5	6	–

Several combinations of parent material class and texture class do not exist (below forests). This is indicated with a "–" in the above table.

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 2 will mainly be correlated with texture classes 2 and 2/3.

APPENDIX 4. LIST OF ADDRESSES

1. Information on and supply of source-receptor matrices:

Dr. Anton Eliassen or
Dr. Trond Iversen
EMEP Meteorological Synthesizing Center-West
Norwegian Meteorological Institute
P.O. Box 43 Blindern
N-0313 Oslo 3
Norway
tel: +47 2 963000
fax: +47 2 963050

2. Information on and supply of the SMART model (dynamic and steady-state) and MACAL model

Ir. W. de Vries
The Winand Staring Center for Integrated Land,
Soil and Water Research
P.O. Box 125
NL-6700 AC Wageningen
The Netherlands
tel. +31 8370 19100
fax. +31 8370 24812

3. Information on and supply of the RAINS model

Dr. Markus Amann
International Institute for applied Systems Analysis (IIASA)
Transboundary Air Pollution Project (TAP)
A-2361 Laxenburg
Austria
tel. +43 2236 71521
fax. +43 2236 71313

4. Information on and supply of the SAFE/PROFILE model

Dr. Harald Sverdrup or
Dr. Per Warfvinge
Dept. of Chemical Engineering
P.O. Box 124
S-221 00 Lund
Sweden
tel. +46 46 107000
fax. +46 46 146030

5. Information and supply of the Water Chemistry Approach

Dr. Arne Henriksen
Norwegian Institute for Water Research
P.O. Box 69 Korsvoll
N-0808 Oslo 8
Norway
tel. +47 2 235280
fax. +47 2 394189

6. Information on and supply of models and methods for uncertainty analysis and mapping in general

Dr. Jean-Paul Hettelingh
Coordination Center for Effects
National Institute of Public Health and Environmental Protection
P.O. Box 1
3720 BA Bilthoven
The Netherlands
tel. +31 30 743529
fax. +31 30 250740
email: mtvhet@rivm.nl

7. Information on and supply of methods for model comparison and software to perform uncertainty analysis

Dr. Robert H. Gardner
Oak Ridge National Laboratory
Environmental Sciences Division
P.O. Box X
Oak Ridge, TN 37931
U.S.A.
tel: +1 615 574 7369
email: gar@ornlsc.bitnet

8. Information on the map of relative sensitivities of ecosystems in Europe to acidic deposition

Stockholm Environmental Institute
Dr. Michael J. Chadwick
Mr. Johan C. I. Kuylensstierna
University of York
York YO1 5DD
United Kingdom
tel. +44 904 432892
fax. +44 904 415185