# Critical loads for eutrophication and acidification for European terrestrial ecosystems

**Final report**

German Environment Agency

**Umwelt Bundesamt**

# Critical loads for eutrophication and acidification for European terrestrial ecosystems

Final report

by

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**Abstract: Critical loads for eutrophication and acidification for European terrestrial ecosystems**

In this final report of the project "Critical loads for eutrophication and acidification for European terrestrial ecosystems" a description is given of the datasets used to construct a database that can be used as a basis for critical load computations. Datasets are described in general terms. Furthermore, the derivation of input data for the critical load models is described in detail. Next, a description is given of an R package and R scripts that can be used for these critical load computations. Both the installation of the scripts as well as their functioning is described and so are the associated data. Thereafter, a 'validation' is given of the R package and R scripts. Results are validated against the 2017 results from the Fortran based background data base computations of RIVM-CCE. Furthermore, a comparison is made with national critical load data submitted to the CCE by Ireland and Germany. Finally, critical loads related to the eutrophying effects of nitrogen are compared to empirical critical loads of nitrogen.

**Kurzbeschreibung: Ökologische Belastungsgrenzen (Critical Loads) für eutrophierende und versauernde Einträge für europäische land-basierte Ökosysteme**

Dieser Endbericht des Projekts „Critical loads for eutrophication and acidification for European terrestrial ecosystems" beschreibt die Datensätze, die verwendet wurden um eine Datenbank zu generieren, die als Grundlage für die Berechnung von Critical Loads genutzt werden kann. Die einzelnen Datensätze werden nur allgemein beschrieben, während die Herleitung der Eingabedaten für die Critical Loads Modelle im Detail beschrieben werden. Im Weiteren werden R-Scripte und ein R Programmpaket beschrieben, die für die Critical Loads Berechnungen verwendet werden können. Sowohl die Installation der R-Scripte als auch deren Funktionsweise werden beschrieben. Des Weiteren wird eine ‚Validierung' der R-Scripte und des R Programmpakets präsentiert. Resultate werden verglichen mit den Resultaten, die 2017 mit einer Fortran-basierten Software vom RIVM-CCE generiert wurden. Auch ein Vergleich wird durchgeführt mit nationalen Critical Loads Daten, die 2017 von Irland und Deutschland zum RIVM-CCCE transferiert wurden. Schlussendlich werden die berechneten eutrophierenden Critical Loads mit empirischen Critical Loads für Stickstoff verglichen.

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## List of abbreviations

| ANC | Acid Neutralizing Capacity |
|---|---|
| Bc | Base cations (Ca+Mg+K) |
| BC | Base cations (Bc+Na) |
| CCE | Coordination Centre for Effects |
| CEC | Cation Exchange Capacity |
| CL | Critical Load |
| CDF | Cumulative Distribution Function |
| EUDB | European Data Base for critical loads |
| EUNIS | European Nature Information System |
| ICP M&M | International Cooperative Programme on Modelling and Mapping |
| LRTAP | (Convention on) Long-range Transboundary Air Pollution |

| ANC | Acid Neutralizing Capacity |
|-----|----------------------------|
| N | Nitrogen |
| RIVM | Rijksinstituut voor Volksgezondheid en Milieu (NL) |
| S | Sulphur |
| SMB | Simple Mass Balance |
| UBA | Umweltbundesamt (German Environment Agency) |
| UNECE | United Nations Economic Commission for Europe |
| WGE | Working Group on Effects (under the LRTAP Convention) |

# Summary

As a result of the observed relationship between air pollution and acidification of soils and waters, the United Nations Economic Commission for Europe (UNECE) initiated in 1979 the Convention on Long-range Transboundary Air Pollution (LRTAP). Under the Working Group on Effects (WGE), the ICP on Modelling and Mapping of Critical Levels and Loads and Air Pollution Effects (ICP M&M) is responsible for the assessment of regional critical loads. Critical loads for acidity and eutrophication are commonly modelled with simple mass balance (SMB) models. In integrated assessments, critical loads are used to compute cost-effective emission abatement measures based on ecosystem vulnerability (expressed by these critical loads) and emission abatement costs, optimized in a European framework. Critical loads are therefore an important input for (European) air pollution policies.

A main task of the Coordination Centre for Effects (CCE), the data centre of the ICP on Modelling & Mapping, is to collect and collate national data on critical loads (CLs), and to provide European maps and databases to the relevant bodies under the LRTAP Convention. For meaningful applications, a complete European coverage with CLs is desirable/required. If a country did not contribute national data, the former CCE at RIVM in the Netherlands filled the gaps with CLs from a so-called European Background Database (EU-DB) of critical loads, which was maintained (and regularly updated) by the CCE in collaboration with Wageningen Environmental Research.

After the CCE was transferred from the RIVM to the UBA in 2018, a revision of the existing background database was required. Therefore, Wageningen Environmental Research was commissioned to develop a database and computational procedures to compute critical loads for eutrophication (by nitrogen) and acidification (by nitrogen and sulphur) for terrestrial ecosystems in Europe.

In the new system, the following critical loads for N and S were computed with the Simple Mass Balance (SMB) method: the maximum critical load for sulphur (CLmaxS), the minimum critical load for nitrogen (CLminN), the maximum critical load for nitrogen (CLmaxN) and the critical load for nutrient nitrogen (CLnutN). CLmaxS can be based on critical values for various chemical criteria such as molar [Al]:[Bc] ratio in soil solution, pH or base saturation.

To compute critical load for (semi-)natural ecosystems, information is needed on ecosystem characteristics such as vegetation cover and soil. We therefore combined six maps to construct a background data base for critical load computations: (1) Land cover, (2) Soil type, (3) Forest growth region, (4) Distance to coast, (5) Natura 2000 delineations, (6) Country borders.  Critical load computations we restricted to (semi) natural habitats, i.e. forests and (semi-)natural vegetation (mires, bogs and fens, natural grasslands and heathland, scrub and tundra).

These maps were gridded in an ArcMap Pro procedure in Python to rasters with a resolution of 0.01° × 0.01° for each country separately. Thereafter, the different layers were combined (overlayed).  There are also two regional data sets used: base cation deposition and meteorological data (temperature and precipitation surplus). Precipitation surpluses were computed using the MetHyd model, which was run for the period 1999-2018 using daily meteorological data.

A set of R procedures (called EUDB 2020) was developed to compute the critical loads for Europe. The software consists of two parts: (a) the BGDB package that holds the basic routines such as transfer functions, conversion functions and the critical load routine and (b) the BGRUN scripts to process the meteorological data, compute the hydrology and the 'MainLoop' that combines all data and scripts to compute critical loads for a set of countries. In 'MainLoop' the following sequence of computations is made:

a. Read the input data from the map overlays on soil types, vegetation type, forest growth region etc.

b. Read in input data from plain ASCII files such as forest growth and soil data

c. Make all the necessary conversions (e.g. compute bulk density from soil characteristics)

d. Prepare the meteorological data for use in MetHyd by, e.g., filling the gaps due to missing data

e. Run the MetHyd model

f. Run the critical load model

Critical loads are computed in 'stripes' of 0.5 degrees latitude. The MainLoop runs from south to north through Europe preparing meteorological data, computing hydrology and critical loads for all receptors in the latitude stripe of 0.5 degrees and between -12 and 42 degrees longitude.

Results from the R procedure have been mapped and compared to the results from the background critical loads computed by RIVM-CCE and reported in the CCE Final Report 2017. Compared to the 2017 results a few changes have been made regarding the computation of critical loads:

1. The software was ported to R

2. The MetHyd model uses daily data for 1999-2018 instead of monthly data 1970-2000

3. The Efiscen forest growth data have been updated to the latest (2016) version

Due to these changes, some minor differences occur in inputs (precipitation surplus) and in critical loads between the new EUDB 2020 and the 2017 results, but the patterns over Europe are mostly (almost) identical.

National critical load data bases provided in 2017 by the Irish and German NFCs have been compared with the CLs for those countries generated by EUDB 2020, with a focus on a comparison of ecosystem areas and the critical loads of N and S. Due to the use of regional maps and differences in ecosystem classification, ecosystem areas deviate between national data and EUDB 2020. Since both Ireland and Germany have used methods for computing critical loads that deviate from the 'standard', also critical loads differ from EUDB. For Ireland critical loads from EUDB 2020 are close to the national ones if we adapt the R software such that it 'mimics' the way the national CLs were computed. For Germany, national critical loads have been

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computed using various criteria (pH, Bsat, Al/Bc, no Al depletion) and various critical values for each of these criteria (e.g. critical base saturation between 3% and 62% and critical pH between 4.08 and 6.2) in many classes. Since the assignment of criteria and their values was based on national maps of soils and vegetation, it was not possible to mimic this in EUDB 2020.

Critical loads for eutrophying nitrogen (CLnutN) from EUDB 2020 were compared to empirical critical loads for N. ClnutN values consist of N uptake, critical N leaching, long-term N immobilisation and N denitrification. Empirical critical N loads are mostly based on observed changes in the structure and functioning of ecosystems in field studies, and relate to unwanted changes in species abundance, -composition and/or -diversity ('ecosystem structure'), or to N leaching, -decomposition or -mineralisation rate ('ecosystem functioning'),

From the above two definitions, it is clear that although both critical loads mainly relate to the eutrophying effects of N, they are conceptually different. The SMB critical load has a strong leaching component: for non-forests for example, net uptake is set to zero, acceptable N immobilisation is a low, constant, value for all EUNIS classes, so N leaching is the main term in the computation of CLnutN. For empirical critical loads, the mentioned changes in species abundance, composition and/or diversity may be caused by N enrichment in the soil organic and mineral phase without resulting in enhanced N leaching. Two runs with EUDB 2020 were made, one with a critical N concentration of 0.2 mg N.l$^{-1}$ for conifers forest and 0.3 mg N.l$^{-1}$ for deciduous forests and semi-natural vegetations (which are the standard values from the Mapping Manual) and a second run with values of 3 mg N.l$^{-1}$ for conifers and deciduous forests and 3.5 mg N.l$^{-1}$ for seminatural vegetations. The latter run used critical values thought to be representative for vegetation changes in Western Europe.

When using the strict values for the critical N concentration, SMB based CLnutN varies mostly between 1-3 and 10-15 kg N.ha$^{-1}$. Compared to empirical critical loads, the median value is mostly still lower than the lower end of the empirical range. Ecosystems that occur in areas with a very high precipitations surplus (and thus a much higher N leaching) can have very high critical loads. When using the higher values for the critical N concentration, median values for CLnutN compare quite well with the average values of the empirical range, especially for forest ecosystems. For some non-forest ecosystems, such as 'Alpine grasslands' and 'Tundras', the median CLnutN is much higher than the upper value of the empirical range. In high precipitation areas, the criterion for the critical N concentration of 2-3 mgN.l$^{-1}$ leads to unrealistically high critical loads.

## Zusammenfassung

Aufgrund des beobachteten Zusammenhangs zwischen Luftverschmutzung (durch Schwefel und Stickstoff) und der Versauerung von Böden und Gewässern rief die United Nations Economic Commission for Europe (UNECE) im Jahre 1979 das Genfer Luftreinhalteabkommen (LRTAP Convention) ins Leben. Als Teil der Working Group on Effects (WGE) unter der LRTAP Convention ist das ICP on Modelling and Mapping of Critical Levels and Loads and Air Pollution Effects (ICP M&M) verantwortlich für die Herleitung und Anwendung von ökologischen Belastungsgrenzen (Critical Loads). Critical Loads (CLs) für versauernde und eutrophierende Einträge werden gewöhnlich mit einfachen Massebilanzmodellen (Simple Mass Balance (SMB) models) berechnet. Integrierte Bewertungen (integrated assessments) werden herangezogen um kosteneffektive Emissionsreduktionen zu ermitteln. Diese basieren auf der Sensitivität von Ökosystemen (ausgedrückt durch deren Critical Loads) und den Kosten von Reduktionsmaßnahmen, optimiert in einem europäischen Zusammenhang. Critical Loads sind daher ein wichtiger Input für die (europäische) Luftreinhaltepolitik.

Eine Hauptaufgabe des Coordination Centre for Effects (CCE), das Datenzentrum des ICP M&M, ist es nationale CL-Daten zu sammeln, daraus europäische CL-Karten, Datenbanken und Statistiken zu generieren und den relevanten Körperschaften der LRTAP Convention zur Verfügung zu stellen. Für sinnvolle Anwendungen sind vollständige CL-Karten wünschenswert bzw. erforderlich. Für Länder, die keine CL-Daten zur Verfügung stellten, wurden die Lücken vom früheren CCE (beim RIVM in den Niederlanden) mit CLs der sogenannten ‚European Background Database' (EU-DB) gefüllt. Diese EU-DB wurde von jenem CCE unterhalten und immer wieder aktualisiert, in Zusammenarbeit mit dem ‚Wageningen Environmental Research' Institut.

Nach dem Transfer des CCE zum UBA im Jahr 2018 wurde eine Revision dieser Datenbank (EU-DB) erforderlich. Daher wurde das ‚Wageningen Environmental Research' Institut beauftragt eine Datenbank und Computerprogramme zu entwickeln, die es erlauben Critical Loads für Eutrophierung (durch Stickstoff, N) und Versauerung (durch N und Schwefel, S) für landbasierte Ökosysteme in Europa zu berechnen.

Mit dieser neuen Software wurden die folgenden Critical Loads mit SMB Modellen berechnet: der maximale CL für Schwefel (CLmaxS), der minimale und maximale CL für Stickstoff (CLminN und CLmaxN) und der CL für eutrophierendes N (CLnutN). CLmaxS kann für verschiedene chemische Kriterien berechnet werden, z.B. molares [Al]:[Bc] Verhältnis in Bodenlösung, pH oder Basensaturation.

Um Critical Loads für Ökosysteme zu berechnen werden Daten über die Charakteristika dieser Ökosystem benötigt, wie z.B. räumliche Ausdehnung der Vegetation sowie Eigenschaften der Böden. Daher wurden die folgenden sechs Karten kombiniert um eine Grundlage für die Berechnung von CLs zu schaffen: (1) Landbedeckung, (2) Bodentyp, (3) Waldwuchsregionen, (4) Abstand zur Küste, (5) Natura 2000 Gebiete, und (6) Staatsgrenzen. Die Berechnung von CLs beschränkte sich auf naturnahe Ökosystem (Habitate), d.h. Wälder, Moore und andere Feuchtgebiete, naturnahes Gras- und Heideland, Buschland und Tundra.

Die Dateninhalte dieser Karten wurden – separat für jedes Land – mit einer ArcMap Pro Prozedur (geschrieben in Python) auf ein Gitter der Dimension 0.01° × 0.01° projiziert und überlappt (kombiniert). Auch zwei weitere, schon gerasterte, europäische Datensätze wurden

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herangezogen: die Deposition basischer Kationen und meteorologische/hydrologische Daten (Temperatur und Niederschlagsüberschuss). Niederschlagsüberschüsse wurden mit dem MetHyd-Modell für die Periode 1999-2018 berechnet, unter Verwendung täglicher meteorologischer Daten.

Um Critical Loads für Europa zu berechnen wurde eine R-Umgebung (genannt ‚EUDB 2020') entwickelt. Diese besteht aus zwei Teilen: (a) dem BGDB Paket, welches die elementaren Routinen (Unterprogramme) enthält, wie z.B. Transferfunktionen, Konversionsfunktionen und die Critical Loads Routine (SMB Modell), und (2) den BGRUN Skripten um die meteorologischen Daten zu verarbeiten, die Hydrologie zu modellieren, sowie den ‚MainLoop' dier alle Daten und Skripte kombiniert zur Berechnung der Critical Loads für die ausgewählten Länder. Im ‚MainLoop' wird die folgende Sequenz von Berechnungen durchgeführt:

a. Einlesen der Daten, die aus der Überlappung der Karten für Bodentyp, Vegetationstyp, Waldwachstumsgebiete usw. gewonnen wurden;

b. Einlesen von Daten von einfachen ASCII-Files, z.B. Waldwachstum und Bodeneigenschaften;

c. Durchführung aller notwendigen Be-/Umrechnungen (z.B. Berechnung der Bodendichte aus Bodeneigenschaften);

d. Vorbereitung der meteorologischen Daten für das MetHyd-Modell; z.B. das Interpolieren von fehlenden Daten;

e. Ausführung des MetHyd-Modells;

f. Ausführung des Critical Load Modells.

Critical Loads werden für alle Rezeptoren, in West-Ost-Streifen mit 0.5° Breite und zwischen -12° und 42° geographischer Länge, berechnet. Der ‚MainLoop' läuft von Süden nach Norden durch Europa und bereitet meteorologische Daten auf, modelliert die Hydrologie und berechnet anschließend die Critical Loads.

Resultate dieser Berechnungen wurden kartiert und mit Critical Loads Daten verglichen, die vom RIVM-CCE berechnet wurden und im CCE Final Report 2017 publiziert sind. Im Vergleich zu den 2017 Berechnungen wurden hier folgende Veränderungen vorgenommen:

1. Die Software wurde in R umgeschrieben

2. Das Methyd-Modell benutzt tägliche meteorologische Daten der Periode 1999-2018 anstatt monatlicher Daten von 1970-2000

3. Die Eficsen Waldwachstumsdaten wurden auf den neuesten Stand (2016) gebracht

Diese Aktualisierungen ergaben einige kleinere Veränderungen in den Inputs (z.B. Niederschlagsüberschuss) und in den Critical Loads, aber die großen Muster über Europa sind beinahe identisch.

Nationale Critical Loads Daten, die 2017 vom irischen und deutschen NFC dem ICP M&M zur Verfügung gestellt worden waren, wurden mit den Daten verglichen, die für diese Länder mit EUDB 2020 generiert wurden, mit dem Fokus auf Vergleiche der Ökosystemfläche und der CLs für N und S. Aufgrund des Gebrauchs europäischer Karten und unterschiedlicher Ökosystem-Klassifikationen divergieren die Ökosystemflächen zwischen diesen nationalen und den EUDB 2020 Daten. Da sowohl Irland als auch Deutschland Berechnungsmethoden benutzten, die vom ‚Standard' abweichen, unterscheiden sich auch deren CLs von den mit EUDB 2020 berechneten. Für Irland werden die EUDB 2020 CLs den national berechneten ähnlich, wenn die R-Software so adaptiert wird, dass es die irische Methode wiederspiegelt. In Deutschland wurden die 2017 CLs mit verschiedenen chemischen Kriterien (pH, Bsat, Al/Bc, etc.) und verschiedenen Werten für diese Kriterien (z.B. kritischer pH zwischen 4.08 und 6.2, kritische Basensaturation zwischen 3% und 62%) für unterschiedliche Boden- und Vegetationsklassen berechnet. Da die Zuordnung der Kriterien und deren Werte auf nationalen Karten basierte, war es unmöglich dies in EUDB 2020 zu realisieren.

Mit EUDB 2020 berechnete Critical Loads für eutrophierenden Stickstoff (ClnutN) wurden mit empirischen Critical Loads für N verglichen. ClnutN-Werte setzen sich zusammen aus N-Aufnahme, N-Ausfluss, langzeitliche Immobilisierung und Denitrifikation. Empirische Critical Loads basieren hauptsächlich auf im Felde beobachteten Veränderungen in der Struktur und Funktion von Ökosystemen. Deren Werte beziehen sich auf unerwünschte Veränderungen in Artenhäufigkeit, Artenzusammensetzung und/oder Diversität (‚Ökosystemstruktur') oder N-Ausfluss, N-Dekompostierung oder N-Mineralisierung (‚Ökosystemfunktion').

Von den zwei obigen Definitionen der CLs ist klar, dass – obwohl sich beide auf eutrophierende Effekte von N beziehen – sie konzeptionell verschieden sind. Der mit SMB berechnete ClnutN wird maßgeblich durch den N-Ausfluss bestimmt: z.B. ist für Nicht-Wälder die Netto-N-Aufnahme null und die akzeptable N-Immobilisierung ein niedriger Wert für alle EUNIS-Klassen, d.h. N-Ausfluss ist der größte Beitrag zu ClnutN. Betreffend empirische CLs, die erwähnten Veränderungen in der Häufigkeit, Zusammensetzung und/oder Diversität der Spezies könnten durch die Anreicherung von N in den organischen und mineralischen Bodenlagen efolgt sein, ohne zu einem erhöhten Ausfluss von N zu führen. Zwei Läufe mit EUDB 2020 wurden gemacht: (a) mit kritischer N-Konzentration von 0.2 mg N.l$^{-1}$ für Koniferen und 0.2 mg N.l$^{-1}$ für alle andere Vegetation (die ‚Standardwerte' aus dem ‚Mapping Manual'), und (b) mit kritische N-Konzentration von 3 mg N.l$^{-1}$ für Wälder und 3.5 mg N.l$^{-1}$ für Nicht-Wälder. Die Werte für den zweiten Lauf werden als repräsentativ für Vegetationsveränderungen betrachtet.

Die Verwendung der strikten Werte für die kritische N-Konzentration ergibt SMB-basierte CLnutN-Werte zwischen 1-3 und 10-15 kg N.ha$^{-1}$. Deren Median ist meistens niedriger als das untere Ende des Intervalls der empirischen Critical Loads. Ökosysteme, die in Regionen mit hohem Niederschlagsüberschuss (und daher hohem N-Ausfluss) vorkommen, können sehr hohe ClnutN-Werte haben. Wenn die höheren Werte für kritische N=Konzentration benützt werden, sind die Medianwerte von ClnutN gut vergleichbar mit den Mittelwerten der empirischen CL-Werte, besonders für Waldökosysteme. Für einige Nicht-Waldökosysteme, z.B. ‚Alpines Grasland' und ‚Tundra', ist der ClnutN-Median viel höher als das obere Ende des empirischen CL Intervalls. In Gebieten mit hohem Niederschlagsüberschuss führt eine kritische N-Konzentration von 2-3 mg N.l$^{-1}$ zu unrealistisch hohen Critical Loads.

# 1   Introduction

As a result of the observed relationship between air pollution and acidification of soils and waters, the United Nations Economic Commission for Europe (UNECE) initiated in 1979 the Convention on Long-range Transboundary Air Pollution (LRTAP). Under this convention a number of working groups were established, to investigate all relevant aspects of air pollution and its effects on ecosystems, crops, human health and materials. Under the Working Group on Effects (WGE), the ICP on Modelling and Mapping of Critical Levels and Loads and Air Pollution Effects (ICP M&M) is responsible for the assessment of regional critical loads. Critical loads for acidity and eutrophication are commonly modelled with simple mass balance (SMB) models. In integrated assessments, critical loads are used to compute cost-effective emission abatement measures based on ecosystem vulnerability (expressed by these critical loads) and emission abatement costs, optimized in a European framework. Critical loads are therefore an important input for (European) air pollution policies.

A main task of the Coordination Centre for Effects (CCE), the data centre of the ICP on Modelling & Mapping, is to collect and collate national data on critical loads (CLs), and to provide European maps and databases to the relevant bodies under the LRTAP Convention, especially for the purpose of integrated assessment. Ideally, all those data are based on national data submissions by National Focal Centres (NFCs) upon a Call for Data issued by the ICP M&M upon request by the WGE. For meaningful applications, a complete European coverage with CLs is desirable/required. Thus, if a country did not contribute national data, the former CCE at RIVM in the Netherlands filled in the gaps with CLs from a so-called European Background Database (EU-DB) of critical loads, which was maintained (and regularly updated) by the CCE in collaboration with Wageningen Environmental Research.

After the CCE was transferred from the RIVM to the UBA in 2018, a revision of the existing background database is required, as a simple reproduction of the existing data base is not the best option, because updates of input data are desirable and because of the fact that UBA decided that the documentation of the background data base is incomplete and the existing software is not easily adjustable. Therefore, Wageningen Environmental Research was commissioned to develop a database and computational procedures to compute critical loads for eutrophication (by nitrogen) and acidification (by nitrogen and sulphur) for terrestrial ecosystems in Europe.

In this report the background data used (e.g., maps, data sets) and the computational rules used to derive input data for the critical load model (e.g., transfer functions between soil type and soil characteristics) are documented. The background data are described in general terms as well as by providing their metadata and data sets that accompany the maps are documented. Each of the input parameters for the critical load model is documented and references to relevant literature is provided.

Next, a description is given of an R package and R scripts that can be used for these critical load computations. Both installation of the scripts as well as the functioning is described and so are the associated data. Thereafter, a 'validation' is given of the R package and R scripts. Results are validated against the 2017 results from the Fortran based background data base computations of RIVM-CCE. Furthermore, a comparison is made with national critical load data submitted to the CCE by Ireland and Germany. Finally, critical loads related to the eutrophying effects of nitrogen are compared to empirical critical loads of nitrogen.

## 1.1    Contents of the report

In chapter 2 the maps that have been used to construct the database with computational units (receptors) is described as well as the procedures applied to process and overlay the various maps. In chapter 3 an overview is provided of all SMB input parameter and the procedures to derive these parameters from (geographical) information.

Chapter 4 describes the setup of the R software to compute critical loads, both in general terms by describing the steps to arrive at critical loads as well as by describing the package that holds all basic functions (BGDB) and the scripts that are used to pre-process data and compute the critical loads in a loop over the modelling domain. Also, the input files are described used by the 'MainLoop' script (see below). In Chapter 5 the installation of the R software is described as well as how to run the script to compute the critical loads and which variables are written out. The correctness of the software is verified by comparing results to results from the original Fortran software. In Chapter 6 some examples of results from the R scripts are provided.

Chapter 7 describes the results of the comparison of the R-based critical loads with the 2017 RIVM-CCE background data base critical loads. Chapter 8 compares national critical load data submitted to the CCE by Ireland and Germany with the new results on a European scale. In Chapter 9, critical loads for the eutrophication effect of N (CLnutN) are compared to empirical critical loads of N.

# 2   Geographical data

In this chapter we provide descriptions of the maps used to construct a database with computation units for critical load computations.

To compute critical load for (semi-)natural ecosystems, information is needed on ecosystem characteristics such as vegetation cover (to compute uptake of nutrients) and soil type (to compute/estimate, for example, weathering rates). We therefore combined six maps to construct a background data base for critical load computations:

► Land cover

► Soil type

► Forest growth region

► Distance to coast

► Natura 2000 delineations

► Country borders

There are also two regional data sets used: base cation deposition and meteorological data (temperature and precipitation surplus). The data on base cation deposition will be discussed in this chapter as well, the description of the meteorological is provided in section 4.3.1.

## 2.1   Land cover

The harmonised LRTAP land cover map (Cinderby et al., 2007) is used, on which land cover is classified according to EUNIS codes (Davies & Moss, 2002).

This original map of 2007 was compiled using existing digital and paper sources including the European Environment Agency (EEA) Corine Land Cover 2000, SEI European Land Cover Map (2002 Revision), FAO Soil Map of the World (1974) and the EEA European Biogeographical regions (2005).

The data have been used to generate classes differentiating between various European Nature Information System (EUNIS) codes (http://eunis.eea.europa.eu/). The dataset contains information down to EUNIS level 3 for specific habitat types.

In 2009 an update of the maps was made (Slootweg et al., 2009); it was extended with a few new EUNIS classes and updated data for a number of countries. The EUNIS codes on the updated map, which was used here, are (Table 1):

**Table 1: EUNIS codes and description**

| EUNIS code | EUNIS description | Numeric code (E3) | Number of 100×100 m$^2$ grid cells |
|---|---|---|---|
| A | Marine habitats | 1000 | 18 |

| EUNIS code | EUNIS description | Numeric code (E3) | Number of 100×100 m² grid cells |
|---|---|---|---|
| A1 or A2 without A2.5 | Littoral rock and other hard substrata or Littoral sediment without Coastal saltmarshes and saline reedbeds | 1102 | 325,070 |
| A2.5 | Coastal saltmarshes and saline reedbeds | 1250 | 301,489 |
| A3 or A4 | Infralittoral rock and other hard substrata or Circalittoral rock and other hard substrata | 1304 | 150,030 |
| A3 or A4 or A5 | Infralittoral rock and other hard substrata or Circalittoral rock and other hard substrata or Sublittoral rock | 1349 | 174,407 |
| A5 | Sublittoral sediment | 1500 | 54 |
| B | Coastal habitats | 2000 | 3,857,073 |
| C1 | Surface standing waters | 3100 | 15,741,695 |
| C1 or C2 | Surface standing waters and surface running waters | 3102 | 45,644,462 |
| C2 | Surface running waters | 3200 | 1,906,137 |
| C3 | Littoral zone of inland surface waterbodies | 3300 | 783,152 |
| D1 | Raised and blanket bogs | 4100 | 7,894,818 |
| D2 or D4 | Valley mires, poor fens and transition mires or Base-rich fens and calcareous spring mires | 4204 | 1,175,174 |
| E1 without E1.2, E1.7, E1.8, E1.9, E1.A | Dry grasslands without Perrenial grasslands and basic steppes or Non-Mediterranean dry acid and neutral closed grassland or Non-Mediterranean dry acid and neutral closed grassland or Mediterranean dry acid and neutral closed grassland or Mediterranean dry acid and neutral open grassland | 5109 | 509,283 |
| E1.2 | Perrenial grasslands and basic steppes | 5120 | 65,872,200 |
| E1.7 or E1.9 | Non-Mediterranean dry acid and neutral closed grassland or Non-Mediterranean dry acid and neutral closed grassland | 5179 | 45,482,750 |
| E1.8 or E1.A | Mediterranean dry acid and neutral closed grassland or Mediterranean dry acid and neutral open grassland | 5189 | 16,559,146 |
| E2 without 2.3 | Mesic grasslands without Mountain hay meadows | 5209 | 131,234,216 |
| E2.3 | Mountain hay meadows | 5230 | 8,527,801 |
| E3 | Seasonally wet and wet grasslands | 5300 | 75,549,849 |
| E4 | Alpine and subalpine grasslands | 5400 | 5,461,568 |

| EUNIS code | EUNIS description | Numeric code (E3) | Number of 100×100 m² grid cells |
|---|---|---|---|
| E5 | Woodland fringes and clearings and tall forb stands | 5500 | 23,862,969 |
| F1 | Tundra | 6100 | 32,184,362 |
| F2 | Arctic, alpine and subalpine scrub | 6200 | 6,999,695 |
| F4 | Temperate shrub heathland | 6400 | 38,091,789 |
| F5 or F6 | Maquis, arborescent matorral and thermo-Mediterranean brushes or Garrigue | 6506 | 12,303,313 |
| F9 | Riverine and fen scrubs | 6900 | 359,039 |
| G1 | Broadleaved deciduous woodland | 7100 | 77,352,602 |
| G1.1 | Riparian Salix, Alnus and Betula woodland | 7101 | 9,232 |
| G1.6 | Fagus woodland | 7106 | 5,750,505 |
| G2 | Broadleaved evergreen woodland | 7200 | 9,398 |
| G2.1 | Mediterranean evergeeen [Quercus] woodland | 7201 | 3,434,898 |
| G3 | Coniferous woodland | 7300 | 130,382,924 |
| G3.1 | Abies and Picea woodland | 7301 | 4,475,944 |
| G3.2 | Alpine  Larix - Pinus cembra woodland | 7302 | 3,296,956 |
| G3.4 | Pinus sylvestris woodland south of the taiga | 7304 | 3,834,720 |
| G3.6 | Subalpine mediterranean Pinus woodland | 7306 | 1,999,355 |
| G4 | Mixed deciduous and coniferous woodland | 7400 | 145,907,408 |
| G4.1 | Mixed swamp woodland | 7401 | 246,007 |
| G4.2 | Mixed taiga woodland with Betula | 7402 | 2,686,294 |
| G4.3 | Mixed subtaiga-taiga woodland with acidiphilous Quercus | 7403 | 348,580 |
| G4.4 | Mixed Pinus sylvestris - Betula woodland | 7404 | 663,516 |
| G4.6 | Mixed Abies - Picea - Fagus woodland | 7406 | 1,532,234 |
| G4.7 | Mixed Pinus sylvestris - acidiphilous Quercus woodland | 7407 | 29,365 |
| G4.B | Mixed mediterranean Pinus - thermophilous Quercus woodland | 7411 | 87 |
| G4.C | Mixed Pinus sylvestris - thermophilous Quercus woodland | 7412 | 15,860 |
| G4.E | Mixed mediterranean pine - evergreen oak woodland | 7414 | 94,852 |

| EUNIS code | EUNIS description | Numeric code (E3) | Number of 100×100 m² grid cells |
|---|---|---|---|
| H | Inland vegetated or sparsely vegetated habitats | 8000 | 138,420 |
| H3 | Inland cliffs, rock pavements and outcrops | 8300 | 6,130,358 |
| H4 | Snow or ice-dominated habitats | 8400 | 4,313,769 |
| H5 | Miscellaneous inland habitats with very sparse or no vegetation | 8500 | 86,018,750 |
| II | Irrigated arable land | 9100 | 519,278,234 |
| IN | Non-irrigated arable land | 9200 | 1,278,061 |
| J | Constructed, industrial and other artificial habitats | 10000 | 24,548,627 |
| Y | Unknown | 25000 | 1,427,273 |

For the critical load database we restricted the EUNIS classes to (semi-)natural habitats, i.e. forests (EUNIS code 'G') and (semi-)natural vegetation ('D': mires, bogs and fens, 'E': natural grasslands and 'F': heathland, scrub and tundra).

## 2.2   Soils

The European Soil Database v2 map (European Soil Bureau Network, 2004) at a scale 1:1 M was used, which includes the whole of Europe and Belarus, the Ukraine and the entire Russian territory. The Soil Database is the result of a EU-JRC project involving all the European Union and neighbouring countries. The methodology used to classify main soil types is based on the terminology of the FAO legend for the Soil Map of the World at scale 1:5,000,000. This terminology has been updated so it can be used for the landscapes in Europa.

The database consists of a list of Soil Typological Units (STU). Soil Topological Units are classified into more than 200 soil types and have attributes that specify the properties of the soil, for example the texture, parent material class and drainage class, etc. The geographical representation was chosen at a scale corresponding to the 1:1,000,000. However, at this scale, it is not feasible to delineate single STUs. Therefore, they are grouped into Soil Mapping Units (SMU) to form soil associations. Each SMU is represented by one or more polygons on the map. The map is accompanied by a table that provides for each SMU the associated STUs and the fraction of the SMU occupied by each of these STUs; but with unknown location of a STU within the SMU.

Six texture classes (including peat) are defined, based on clay and sand content (FAO-UNESCO, 2003); Table 2. The drainage classes, which are used to estimate the denitrification fraction, were derived from the dominant annual soil water regime (FAO-UNESCO, 2003).

**Table 2: Texture class definition**

| Soil texture code | Soil texture definition |
|---|---|
| 0 | No information |

| Soil texture code | Soil texture definition |
|---|---|
| 9 | No mineral texture (Peat soils) |
| 1 | Coarse (18% < clay and > 65% sand) |
| 2 | Medium (18% < clay < 35% and >= 15% sand, or 18% < clay and 15% < sand < 65%) |
| 3 | Medium fine (< 35% clay and < 15% sand) |
| 4 | Fine (35% < clay < 60%) |
| 5 | Very fine (clay > 60 %) |

## 2.3   Forest growth regions

Forest growth regions for Europe were taken from the EFI database (Schelhaas et al., 2006) that provides data for about 250 regions in (most of) Europe for various species and age classes. Forest growth was regionally modelled using the EFISCEN model based on the observed growth curves from the EFI data base (Petz et al., 2016; Prins et al., 2017).

For the parts of Russia mapped, we used the forest regions from Alexeyev et al. (2004), who compiled data for 74 administrative regions within Russia.

## 2.4   Distance to coast

The distance to coast is used for deriving base cation deposition for those areas where no EMEP estimates of base cation deposition are available; it was taken from a 2009 NASA dataset.

The data set was generated at a global grid increment of 0.04 degrees and then interpolated to a 0.01-degree resolution and saved the as a GeoTiff image.

(see https://oceancolor.gsfc.nasa.gov/docs/distfromcoast/).

## 2.5   Natura 2000 areas

Critical loads are of particular interest for nature protection areas. For EU assessments the European Union's Natura 2000 (N2k) areas were integrated into the EU-DB.  The borders of the Natura 2000 areas from 2019 can be found at https://www.eea.europa.eu/data-and-maps/data/natura-10. "Natura 2000 is a network of core breeding and resting sites for rare and threatened species, and some rare natural habitat types which are protected in their own right. It stretches across all 28 EU countries, both on land and at sea. The aim of the network is to ensure the long-term survival of Europe's most valuable and threatened species and habitats, listed under both the Birds Directive and the Habitats Directive" (see https://ec.europa.eu/environment/nature/natura2000/index_en.htm).

## 2.6   Base cation deposition

Base cation (BC) deposition for Europe was obtained from an atmospheric dispersion model for BC (Van Loon et al., 2005) for the EMEP 50×50 km grid. For areas in Eastern Europe for which Van Loon et al. (2005) does not provide depositions, so in these areas in EUDB 2020 calcium (Ca) deposition was taken from a global map computed with a model of Tegen and Fung (1995). Comparing the spatial patterns of Ca deposition of the European map and the global map, it is clear that the global Ca map underestimates the deposition in Europe by at least a factor of two.

This underestimation was also recognized by Lee and Pacyna (1999) who attribute the difference mainly to the fact that they did not include all important, local, sources of Ca in their modelling. Using both maps would thus lead to a non-smooth transition in BC deposition in eastern Europe. Since the European map does include all sources (natural and anthropogenic) it was taken as the reference and the Ca from the global were multiplied by two to generate a consistent deposition pattern over the entire modelling area in the combined map.

Magnesium (Mg) and potassium (K) deposition are also needed; relationships between Ca deposition and Mg and K deposition were derived on the basis of measurements at 95 EMEP/CCC monitoring stations in Europe (Hjellbrekke & Tarrason, 2001). Because of the different origin of base cations, the spatial patterns in these ratios are far from constant. In southern areas Bc input is dominated by Ca from Saharan dust, whereas in Northern Europe Mg and K become more important with Mg dominating the Bc input in coastal regions. In EUDB 2020 Mg deposition was modelled as a function of Ca deposition, $Ca_{dep}$ (eq.ha$^{-1}$.yr$^{-1}$), and the distance to coast based on a regression on the basis of measurements at 95 EMEP/CCC monitoring stations in Europe (Hjellbrekke & Tarrason, 2001) and the distance to coast of the monitoring station:

(2.1) $\quad Mg_{dep} = a \cdot Ca_{dep} + b$

Where $a$ is a constant and $b$ depends on the distance to the coast. Regression (r$^2$=0.375) yields $a$=0.4748; values for $b$ are listed in Table 3 for the various distance-to-coast classes.

**Table 3: Regression coefficient for computing Mg deposition**

| Distance to coast (km) | Regression coefficient $b$ |
| --- | --- |
| <10 | 239.6 |
| 10-20 | 171.6 |
| 20-50 | 104.9 |
| 50-100 | -18.2 |
| >100 | -39.4 |

Mg deposition thus increases with increasing Ca deposition and strongly decreases with distance from the coast.

K deposition was also estimated as a function of Ca deposition, $Ca_{dep}$ (in eq.ha$^{-1}$.yr$^{-1}$), and latitude, $Ylat$ (in degrees) using a regression on the data from the EMEP/CCC monitoring stations:

(2.2) $\quad K_{dep} = a + b \cdot Ca_{dep} + c.Ylat$

Regression yields $a$=−95.1, $b$=0.2419 and $c$=1.731 with r$^2$ = 0.552; K deposition increases with increasing Ca deposition and with latitude. For each receptor outside the domain of the van Loon data set, Mg and K deposition was computed using these two equations on the basis of their latitude and distance to coast. For the few areas in eastern Europe for which Van Loon et al. (2005) does not provide depositions, the deposition of natrium (Na) and chlorine (Cl) where assumed to be of equal magnitude and set to zero; the magnitude of the values has no influence on the computed critical load as long as both values are equal, because neither Na nor Cl is taken up by the vegetation.

Since S-depositions from EMEP do not include natural emissions from the sea, base cation and chloride depositions in eq.3-2 are sea-salt corrected, assuming that all Cl originates from sea salts (for the methodology see a.o. the Mapping Manual (CLRTAP, 2017).

## 2.7    Meteorological data and hydrology

To compute the leaching flux of ions from the rooting zone and to estimate the temperature effect on weathering and some chemical parameters, a set of meteorological data and a model that computes leaching fluxes is needed. Both the meteorological data sets are described in section 3.2.1.

## 2.8    Overlay procedure

### 2.8.1    General procedure

These five maps were gridded in ArcMap to rasters with a resolution of 0.01°× 0.01° for each country separately. Thereafter, the different layers were combined (overlayed).  Then, within the R scripts, grids with common soil, vegetation and region characteristics can be merged within blocks of e.g. 0.10° × 0.05°. This means that all receptors with equal attributes (soil, land use etc.) are grouped and their areas summed to reduce the number of computations for receptors which are identical in their critical load inputs.

### 2.8.2    Details of the ArcGIS procedures

A python script was made to process the basic data described in section 2.1 to 2.5. The script is stored in an ArcGIS toolbox. Opening this toolbox, and running the data-processing script in ArcGIS displays a menu in which the user can specify input and output of the overlay procedure (Figure 1).

**Figure 1: Input form for the ArcGIS procedure to process the soil data**



In this input form the user must define the basic input shapes or rasters (soils, administrative regions (countries), land use, Efiscen regions, N2000 regions and distance to coast), a file that

defines the relationship between Soil Mapping Units and Soil Typological Units, and the file with the definition of the EUNIS classes in the landuse maps and finally the location of the output geodatabases.

The data processing Python script exists of six processing blocks:

**Block 1: Soil map processing**

# process 1: Creates two file GDB's at a specified location (Output folder location). These databases are named by default; 'soils.gdb', 'metadata.gdb'.

# process 2: Table to table; stores the .csv file containing the SMU_STU relationship in the soil.gdb.

# process 3: Process SMU table; add the STU information (soil type, relative area) to the SMUs.

# process 4: Create a geodatabase table for the shapefile.

# process 5: Join the shape with the newly constructed attribute table with both SMU and STU information.

# process 6: Feature Class to Geodatabase; store the shapefile and its attributes in the geodatabase

# process 7: Feature to Raster conversion; creates a raster from the shapefile on 0.01 x 0.01 degrees resolution.

# process 8: Create separate country files by country code; split the raster into rasters per country based on the geodatabase with country borders. Generate soil rasters per country.

**Block 2: Land use map processing**

# process 1: Creates a directory named 'landuse' if it does not yet exist.

# process 2: Creates a file GDB named 'landuse'.

# process 3: Convert ASCII to raster; converts all ASCII files to rasters and renames the output files with the country codes used by GADM36_0. No data values within the rasters are replaced by a value of -9999.

**Block 3: EFISCEN processing**

# process 1: Create directory if it does not yet exist and a file GDB both named 'efiscen'.

# process 2: creates Efiscen raster files per country using the country code.

**Block 4: Natura 2000**

# process 1: Create a file GDB named 'N2000'.

# process 2: Projects the dataset to EPSG 4326, WGS84

# process 3: Converts the N2000 feature dataset to a raster dataset and subsequently generates grids per country using the country codes.

**Block 5: Distance to coast**

# process 1: Creates a file GDB named 'Dist2Coast'.

# process 2: Extract by rectangle; extracts the right map extent from the 'Distance to Coast input file'

# process 3: Creates 'Distance to Coast' grids per country using the country codes.

**Block 6: Combine grids**

# Process 1: Creates a directory named 'combined' in the Output folder location to store the final .tif files.

# Process 2: Combines 'Soil', 'Landuse', 'efiscen', 'Natura 2000' and 'Dist2Coast' grids.

# Process 3: Removes redundant information.

The progress of the script is shown in a progress window:

**Figure 2: Progress window**
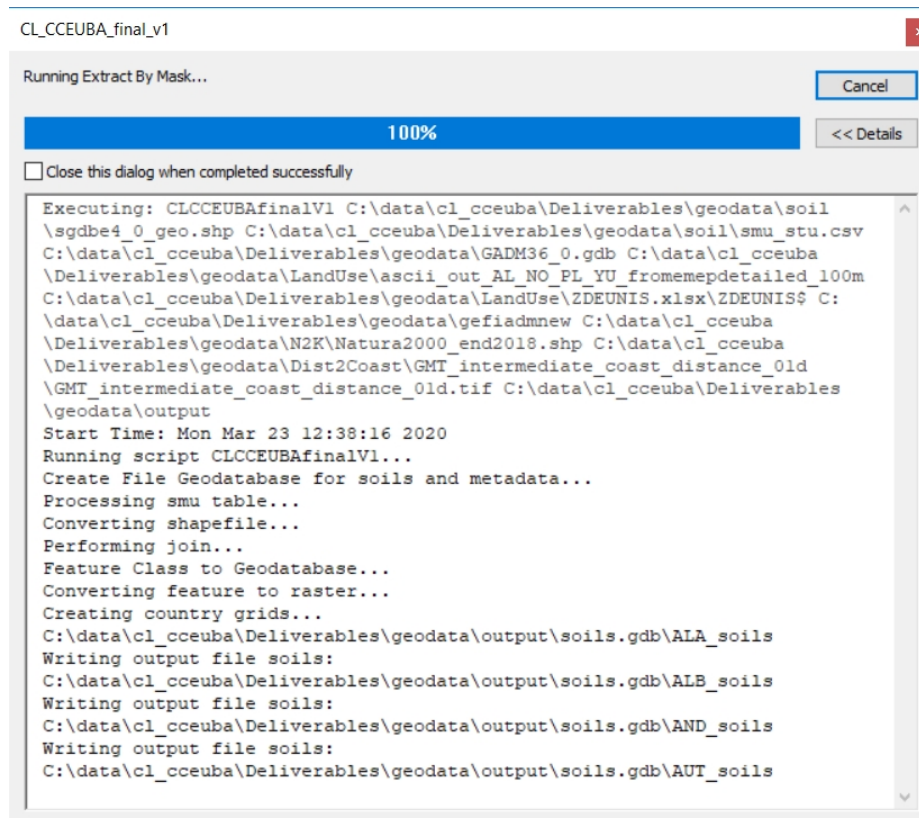


After all basic grids and shapefiles have been processed this way, the country specific grids are combined and an export is made to geo-referenced tiff files with attribute data that can be read by the R procedures that computes the critical loads.

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# 3 Input data for the critical load model

## 3.1 Simple Mass Balance (SMB) critical load equations

### 3.1.1 Introduction

Critical loads for N and S can be computed with the Simple Mass Balance method. According to the Mapping Manual (CLRTAP, 2017): "The purpose of a model-based approach to calculating critical loads is to link, via mathematical equations, a chemical criterion (critical limit) with the maximum deposition(s) 'below which significant harmful effects on specified sensitive elements of the environment do not occur', i.e. for which the criterion is not violated." The method has a number of basic assumptions and some simplifications which are extensively discussed in section V3 of the Mapping Manual and in Posch et al. (2015).

### 3.1.2 The critical load of nutrient N

Starting from the mass balance of total N and making a few simplifying assumptions (e.g. assuming complete nitrification of ammonium; for a complete list see section V3.1.1/V3.1.2 of the Mapping Manual), the critical load of nutrient N (eutrophication), $CL_{nut}N$, is obtained as:

$$(3.1) \quad CL_{nut}N = N_i + N_u + \frac{Q \cdot [N]_{acc}}{1 - f_{de}}$$

where $N_i$ is the long-term net immobilisation of N in the soil, $N_u$ is the net removal of N in harvested vegetation, $f_{de}$ ($0 \le f_{de} < 1$) is the fraction of the net N input denitrified, $Q$ is the precipitation surplus (runoff) leaving the soil compartment (rooting zone), and $[N]_{acc}$ is the acceptable (critical) N concentration to avoid 'harmful effects' on the chosen 'sensitive element of the environment'.

Note that a critical load of eutrophication can also be given/determined by a so-called empirical CL, CLempN (see Chapter 8). If both are given for the same ecosystem, the minimum is taken and called CLeutN, i.e. CLeutN = min{CLnutN,CLempN}.

### 3.1.3 Critical loads of N and S acidity

Starting from the charge balance in the soil leaching flux and inserting the (simplified, steady-state) mass balances for S, N, chloride (Cl) and base cations (Bc=Ca+Mg+K, BC=Bc+Na), we obtain as the maximum critical load of S, $CL_{max}S$:

$$(3.2) \quad CL_{max}S = BC_{dep} - Cl_{dep} + BC_w - Bc_u - ANC_{le,crit}$$

where the subscripts *dep* and *u* refer to deposition and net uptake, resp., $BC_w$ is the weathering of base cations, and $ANC_{le,crit}$ the critical ANC (acid neutralising capacity) leaching (see below). More details on the method and the assumptions made can be found in section V3.2.1 of the Mapping Manual. For N, two CLs are defined: the minimum CL of N, $CL_{min}N$, and the maximum CL of N, $CL_{max}N$, derived as:

$$(3.3) \quad CL_{min}N = N_i + N_u \quad \text{and} \quad CL_{max}N = CL_{min}N + \frac{CL_{max}S}{1 - f_{de}}$$

The three quantities $CL_{min}N$ and $CL_{max}N$ and $CL_{max}S$ define the so-called *critical load function* (CLF; see Figure V.3 in the Mapping Manual); and every deposition pair ($N_{dep}$,$S_{dep}$) lying on the CLF is a critical load of acidifying N and S.

## 3.2    Parameters for computing ANC (Acid Neutralising Capacity)

In SMB, the ANC leaching is computed as (Mapping Manual V3.1.2/V3.2.4):

(3.4)     $ANC_{le} = -H_{le} - Al_{le} + HCO_{3,le} + Org_{le}$

with $H_{le}$ = $Q[H]$ with $Q$ being the water flux at the bottom of the root zone, and the other three terms (aluminium, bicarbonate and organic acids) are computed as function of $[H]$.

### 3.2.1    Aluminium

The concentration of aluminium is computed according to:

(3.5)     $[Al] = K_{Alox} \cdot [H]^a$

with an equilibrium constant $K_{Alox}$ and an exponent $a$. The exponent $a$ was set to 3, equivalent to gibbsite equilibrium. The constant $K_{Alox}$ were set to 9.669 for texture class 1, 9.609 for texture class 2  and 8.873 for texture classes 3,4 and 5. These values were derived from a calibration and validation study with VSD on about 80 intensively monitored forest sites where soil solution measurements were available (Reinds et al., 2008b).

### 3.2.2    Bicarbonate

The concentration of bicarbonate is computed according to:

(3.6)     $[HCO_3] = \dfrac{K_1 \cdot K_H \cdot p_{CO2}}{[H]}$

where $K_1$ is the first dissociation constant, $K_H$ is Henry's constant and $p_{CO2}$ is the partial pressure of $CO_2$ in the soil solution. Both the values of $K_1$ and $K_H$ are temperature dependent and computed according to (Harned & Davis, 1943):

(3.7a)     $lgK_H = \dfrac{2386}{T_{abs}} + 0.0153 \cdot T_{abs} - 14.018$

(3.7b)     $lgK_1 = \dfrac{-3404}{T_{abs}} - 0.0328 \cdot T_{abs} + 14.844$

With $T_{abs}$ is the temperature in K. Also the partial pressure of $CO_2$ in soil solution is computed as a function of temperature (after Gunn and Trudgill (1982); see also Mapping Manual):

(3.8)     $log_{10}P_{CO2} = 0.031 \cdot \vartheta - 2.38$

where $\vartheta$ is the temperature in degrees Celsius.

### 3.2.3    Organic acids

The concentration of organic anions is computed from DOC according to the following simple monoprotic model:

(3.9)     $[Org] = \dfrac{m \cdot DOC \cdot K_{org}}{K_{org} + [H]}$

where $DOC$ is the concentration of dissolved organic carbon (in molC m$^{-3}$), $m$ is the concentration of functional groups ('charge density', in mol.molC$^{-1}$) and $K_{org}$ the dissociation constant. The value of $m$ is set to 0.023 mol.molC$^{-1}$ (Santore et al., 1995). The value of $K_{org}$ is set at 4.5 (see the Mapping Manual). The concentration of dissolved organic carbon ($DOC$; in mgC.L$^{-1}$) is estimated from soil solution measurements in the ICP Forest Intensive Monitoring data as a function of texture class and a 'characteristic' soil pH:

(3.10)     $DOC = max\{54.02 + a_{tex} - 6.67 \cdot pH, 0.\}$

where the texture-dependent constants $a_{tex}$ are given in Table 4; $DOC$ in molC.m$^{-3}$ is obtained by dividing by 12.

**Table 4: Texture class (see Table 2 for definitions) dependent constants for estimating DOC**

| Texture class | | | |
|---|---|---|---|
| **1** | **2** | **3** | **≥ 4** |
| $a_{tex}$    0 | -12.7 | -8.1 | -11.8 |

Soil pH was obtained by calculating median values per soil type and soil group based on the ICP Forest level I monitoring data base (Vanmechelen et al., 1997). Results are provided in Appendix A.1.

## 3.3 Soil parameters

A number of soil parameters is needed to compute critical loads (see section 3.1):

► Thickness of the rooting zone

► Denitrification fraction

► Weathering rates of base cations

► Nitrogen immobilisation

### 3.3.1 Thickness of the rooting zone

The thickness of the rooting zone can be specified by the user in the R script; generally, a value of 0.5m is used representing the thickness of the rooting zone in forests. For lithosols, values are set to 0.1m as these soils are by definition shallower than 0.1m.

### 3.3.2 Denitrification fraction

Dominant water regimes linked to STUs and associated denitrification fractions are listed in Table 5 (modified from Reinds et al. (2001)). These denitrification fractions are based on data by Breeuwsma et al. (1991) for peat, clay soils and sandy soils in the Netherlands. For very dry soils (water regime code 1), the denitrification was set to a (low) value of 0.1.

**Table 5: Dominant annual water regimes**

| Water regime code | Water regime description | Denitrification fraction |
|---|---|---|
| 0 | No information | 0.2 |
| 1 | Not wet within 80 cm for over 3 months, nor wet within 40 cm for over 1 month | 0.1 |
| 2 | Wet within 80 cm for 3 to 6 months, but not wet within 40 cm for over 1 month | 0.4 |

| Water regime code | Water regime description | Denitrification fraction |
|---|---|---|
| 3 | Wet within 80 cm for over 6 months, but not wet within 40 cm for over 11 months | 0.5 |
| 4 | Wet within 40 cm depth for over 11 months | 0.8 |

### 3.3.3 Weathering rates of base cations

The weathering rates of base cations were computed according to the Mapping Manual (section V3.2.3.3). Each soil type was assigned a parent material class first. This parent material class was derived from the soil map (it is an attribute to each STU); in the few cases the soil map does not provide a parent material class for the STU, it was based on the general relationship between soil type and parent material class listed in Table V.15 of the Mapping Manual. Based on soil texture (also an attribute to each STU) and parent material class, a weathering rate class was assigned to each STU according to Table V.16 of the Mapping Manual. The actual weathering rate of base cations (in eq.m$^{-2}$ for a soil of depth $z$) was then computed according to (Mapping Manual, equation V.44):

$$(3.11) \qquad BC_{we} = z \cdot 0.05 \cdot (iwcl - 0.5) \cdot 10^{\left(\frac{facH}{281} - \frac{facH}{273+T}\right)}$$

with *iwcl* being the weathering rate class, *facH* a constant of 3600 K and *T* the temperature at the site (in degree C). For calcareous soils, weathering rate was set to an (arbitrary) high value of 1 eq/m$^2$ (for a soil of 0.5 m depth). The computed weathering rates were split to weathering rates per base cation (Ca, Mg, K, Na) according to Table 6 (*clay*: clay content, *silt*: silt content of the soil):

**Table 6: Weathering rates of separate base cations as a fraction of the total weathering rate**

| Soil texture class | Ca | Mg | K | Na |
|---|---|---|---|---|
| 1 : sandy soils [1] | 0.3 | 0.3 | 0.2 | 0.2 |
| 3 : loess soils [2] | 0.34*clay+ 0.07*silt-6.19 | 0.08*clay-0.17 | 0.032*clay+ 0.037*silt-2.18 | 0.036*silt-1.55 |
| 2,4,5 : clay soils [2] | 1.60*clay-19.5 | 0.70*clay- 7.0 | 0.06*clay- 0.43 | 0.05*clay+ 0.2 |

   1)    De Vries (1994);  2)  Van der Salm (1999)

### 3.3.4 Nitrogen immobilization

The long-term net N immobilization was set at 0.5 kg N ha$^{-1}$ yr$^{-1}$, which is at the upper end of the estimated annual long-term accumulation rates for Swedish forest soils (Rosén et al., 1992). An overview of current and long-term immobilisation rates is provided by Höhle and Wellbrock (2017), who found both negative as well as highly positive current N immobilisation rates, but conclude that long-term N immobilization ranged from 0.035 to 1.6 kg ha$^{-1}$ yr$^{-1}$ with a median of about 0.2 kg ha$^{-1}$ yr$^{-1}$ for France and Germany and 0.4kg ha$^{-1}$ yr$^{-1}$ for Switzerland which is consistent with the values by (Rosén et al., 1992).

## 3.4 Uptake of nitrogen and base cations

It is assumed that both stems and branches are removed by tree harvesting. The net *potential* growth uptake $X_{upot}$ (in eq.ha$^{-1}$.yr$^{-1}$) of N and base cations is computed as (Mapping Manual Section V3.1.3.2/V3.2.3.4):

(3.12) $\quad X_{upot} = G_{st} \cdot (ctX_{st} + f_{brst} \cdot ctX_{br}), \ X = N, Ca, Mg, K$

where $G_{st}$ is the annual growth rate of stems, $ctX_{,st}$ and $ctX_{,br}$ the elements contents of ion X in stems and branches, resp., and $f_{brst}$ the branch-to-stem ratio of the tree species. This equation assumes that stems and branches grow at the same rate. The information on the tree species (conifers, deciduous, mixed) follows from the EUNIS class on the landcover map.

The net *actual* nutrient uptake in SMB is computed by matching demand and supply. For each base cation *X*, the supply ($X_s$) is computed according to:

(3.13) $\quad X_s = X_{we} + X_{dep} - X_{min} \cdot Q_{le}$

Where the subscript *we* stands for weathering and *dep* for deposition. $X_{min}$ is the minimum concentration of base cation *X* in the leaching water (set at a very low value of $10^{-6}$ eq.m$^{-3}$) and $Q_{le}$ the leaching flux from the root zone (m.yr$^{-1}$). Subsequently, for each ion, the ratio between supply and demand is computed and the minimum of the these four ratios is used to compute the actual uptake $X_{uact}$ for all base cations (assuming that the uptake is reduced for all base cations based on the one with the lowest supply):

(3.14) $\quad X_{uact} = r_{min} \cdot X_{upot}$

with $r_{min}$ being the minimum ratio of supply and demand.

Nitrogen uptake is computed the same way, but a surplus uptake of up to 20% is assumed as long as the resulting multiplier is ≤ 1.0.

The element contents in stem wood and branch wood are taken from the literature review by (Jacobsen et al., 2003). Forest growth in EU countries was taken from the European Forest Information Scenario (EFIscen) model (Schelhaas et al., 2007). Forest growth for the rest of Europe was derived from the EFI database (Schelhaas et al., 2006). Forest growth for Russia was estimated from data by Alexeyev et al. (2004) who compiled statistical data on growing stock and areas of stocked land from available data sources (for more details see Reinds et al. (2008a)). The land cover map distinguishes only between coniferous, broad-leaved (deciduous) and mixed forests. EFIscen, on the other hand, distinguishes 7 species; these are assigned to these 3 forest categories, and the uptake of these categories is computed using the area-weighted average growth of the assigned species (area per species for each region is provided by EFIscen). The net uptake for non-forests is set to zero, because it is assumed that no harvesting takes place, i.e. all nutrients are recycled.

# 4   Methodological approach for computing critical loads

In this chapter we provide a description of the general setup of the R procedures and the package that can be used to compute critical loads.

The software consists of two parts: (a) the BGDB package that holds the basic routines such as transfer functions, conversion functions and the critical load routine and (b) the BGRUN scripts to process the meteorological data, compute the hydrology and the 'MainLoop' that combines all data and scripts to compute critical loads for a set of countries.

## 4.1   Workflow for computing critical loads

The computation of critical loads consists of a set of (preparatory) steps:

1. Pre-process the meteorological data and store the data in .rds files (see section 3.2.1)
2. Run the MainLoop.R script that
   a. Reads the input data from the map overlays on soil types, vegetation type, forest growth region, etc.
   b. Reads in input data from plain ASCII files such as forest growth and soil data
   c. Makes all the necessary conversions (e.g. compute bulk density from soil characteristics)
   d. Prepares the meteorological data for use in MetHyd by, e.g., filling the gaps due to missing data
   e. Runs the MetHyd model
   f. Runs the critical load model

Critical loads are computed in 'stripes' of 0.5 degrees latitude. The MainLoop runs from south to north through Europe preparing meteorological data, computing hydrology and critical loads for all receptors in the latitude stripe of 0.5 degrees and between -12 and 42 degrees longitude.

## 4.2   The BGDB package

The BGDB (BackGroundDataBase) package holds a set of R routines for reading input data, applying (soil) transfer functions and implementing critical load equations. It mimics the setup of the original Fortran Background Database software of the RIVM-CCE. The package contains the following set of routines:

**Table 7: R routines in the BGDB package**

| Name of the R routine | | | | |
|---|---|---|---|---|
| argridd.R | Falbedo.R | Intcosz.R | RegGrown.R | sunset.R |
| BcNPcont.R | fKHCO3.R | Ions.R | SeaSalts.R | trunclin.R |
| claysilt.R | FlgKAlox.R | llemep.R | SMB.R | uptake.R |
| contBcNP.R | fOliver.R | MualvGen.R | snowam.R | weasplit.R |
| coslin.R | F_DOC.R | pedo_rho.R | snowamV.R | wrate.R |
| Daycosz.R | get1BCdep.R | petaet.R | soilchar.R | wratecl.R |
| declinat.R | getKAlox.R | petuv.R | soilMoisture.R | |
| EfromLog.R | get_inp.R | RegGrow.R | SolConst.R | |

A full documentation of each routine can be found in the separate Appendix on BGDB. The naming of the routines follows the original naming of the Fortran routines of the RIVM-CCE.

## 4.3   The BGRUN scripts

The BGRUN (RUNning the computations for the BackGround data base) scripts consist of a stand-alone script to pre-process the meteorological data and of a number of scripts called from the MainLoop script. These are described below. The files related to the BGRUN scripts, are stored in subdirectories of the BGRUN main directory as follows:

**Table 8: Files and directories for the BGRUN script**

| Subdirectory | Contains |
| --- | --- |
| SoilData | the results of the map overlay procedure, 8 files per country in a subdirectory named after the ISO 2-letter code of the country |
| PreData | *File _Efiscen_2020.csv* (Efiscen data) |
| | *File BCTotAve.dep* (Base cation deposition for the EMEP grid) |
| | *File TotalCaS.asc* (total Ca deposition for cells of 1×1 degree) |
| | *File _SoilChar.Tab* (soil data on pH, organic C etc.) |
| | *File _Scode_1M_EAw.Tab* (the relationship between SMU and STU and the STU's soil characteristics (texture etc.) |
| | *File _RegGrow.Tab* (forest growth for the non-Efiscen regions in Europe) |
| | *subdirectory* HydPrep (with the results from the script *MetHyd.R* per 0.5° latitude stripe) |
| | *subdirectory* MetPrep (with the selected meteorological data for MetHyd per 0.5° latitude stripe; results of the script *MkCrudeMet.R, see section 4.3.1*) |
| | *subdirectory* MetGlobs (with the global long-term average monthly meteorological data used for gap-filling, downloaded see section 5.1) |
| | *subdirectory* MetCrops (with the extracted meteorological data from the netcdf files per 0.5° latitude stripe; results of the script cropMetNC.R, see section 5.2) |
| | *subdirectory* Metncs (with the EObs netcdf files with daily data; downloaded see section 5.1) |
| Output | the results of the critical load computations of the R scripts |
| Scripts | all scripts needed for the critical load computations that are not in the BGDB package; See section 5.1-5.3 where these scripts and their functioning is discussed |
| Data | R data files from pre-processing |
| Vignettes | the generated vignettes |

### 4.3.1   Meteorological data for the MetHyd model

Before critical loads can be computed, the hydrology at each site must be known: leaching of nutrients is one of the terms in the critical load equations. In the RIVM-CCE setup, the MetHyd

model (Bonten et al., 2016) was used to compute the leaching flux from the rootzone. For this project, MetHyd was converted from Fortran to R (see script MetHyd.R). The R script was tested and proved to give identical results as the Fortran routines at equal inputs. For the RIVM-CCE background data base, long-term (1961–90) average monthly temperature, precipitation and cloudiness for MetHyd were derived from a high-resolution global database (New et al., 2002) that contains monthly values for the years 1901–2001 for land-based grid-cells of 10' × 10' (approx. 15 km × 18 km in central Europe) that were transformed into daily data for use in MetHyd.

The new R scripts use time series of daily data from the EObs data set basis for the period 1999-2018 (20 years) thus incorporating variability in weather between years and avoiding the need for deriving daily values from monthly data. Furthermore, the EObs data set contains radiation data, instead of cloudiness. However, the EObs data set contains global radiation data whereas we need the net radiation (earlier computed from cloudiness) to the ground. A method has been developed to convert the global radiation data from EObs into net radiation at the ground which is described in Appendix A3.

EObs data files are available as netcdf files through the EObs website (see chapter 3). Before the data can be used, two steps are needed:

The netcdf files need to be processed before they can be used in R: we need to extract the proper time period and store these data in R data frames, one for each stripe of 0.5 degrees latitude using the script *MkCrudeMet.R*. The script can be run from R studio (see Chapter 5). Results are stored in the subdirectory BGRUN\Predata\MetPrep; naming is according to S*XX.X*.rds with *XXX* being the lower value of the latitude stripe in degrees (e.g. S48.5.rds stores all extracted data for the latitude stripe between 48.5 and 49° N and -12 and 42° E).

Data can be missing because gaps occur in the EObs time series or the receptor lies outside the area covered by the EObs data set. To make full timeseries for each location the script *makeMetSlice8.R* is used. Missing values for rain and temperature in the EObs dataset are an exception. We aggregated the data to grids of 0.5° × 0.5° for reasons of computational speed because we analyzed the data and found that the distribution of average rain/temperature/radiation is fairly similar over the 0.1° × 0.1° EObs data cells within the 0.5° × 0.5° grid cells (see Figure 3 for an example for three selected cells in Europe). The aggregation to the 0.5° × 0.5° grid cells is done by selecting the best of the constituent 25 cells. Best is defined by having the maximum number of data rows for the entire time period 1999-2018. Radiation data is often missing; missing values are replaced by using a generalized additive model (which in included in the makeMetSlice8.R script) that estimates radiation on the basis of temperature, rain and day-of-year (DOY) within the latitude stripe based on the available data in the stripe. In makeMetSlice8.R for the receptors that lie in the area not covered by EObs, monthly data from New et al. (2002) are used as a starting point. For each month a nearest neighbor cell from the EObs dataset, in terms of monthly rain and monthly average temperature in the same latitude stripe, is copied as daily meteo. Radiation is modeled with the same model as for the missing data in the EObs region. In the odd case that also these monthly data is missing, rain and temperature data of preceding/succeeding month and identical month in the preceding/succeeding year are averaged.

**Figure 3a: Comparison of the cumulative distribution of daily values for rainfall  (upper 6 graphs) and rainfall (lower six graphs) for 4 months (1,4,7,10) at 2 locations in 3 selected 0.5 × 0.5˚ cells in Europe**
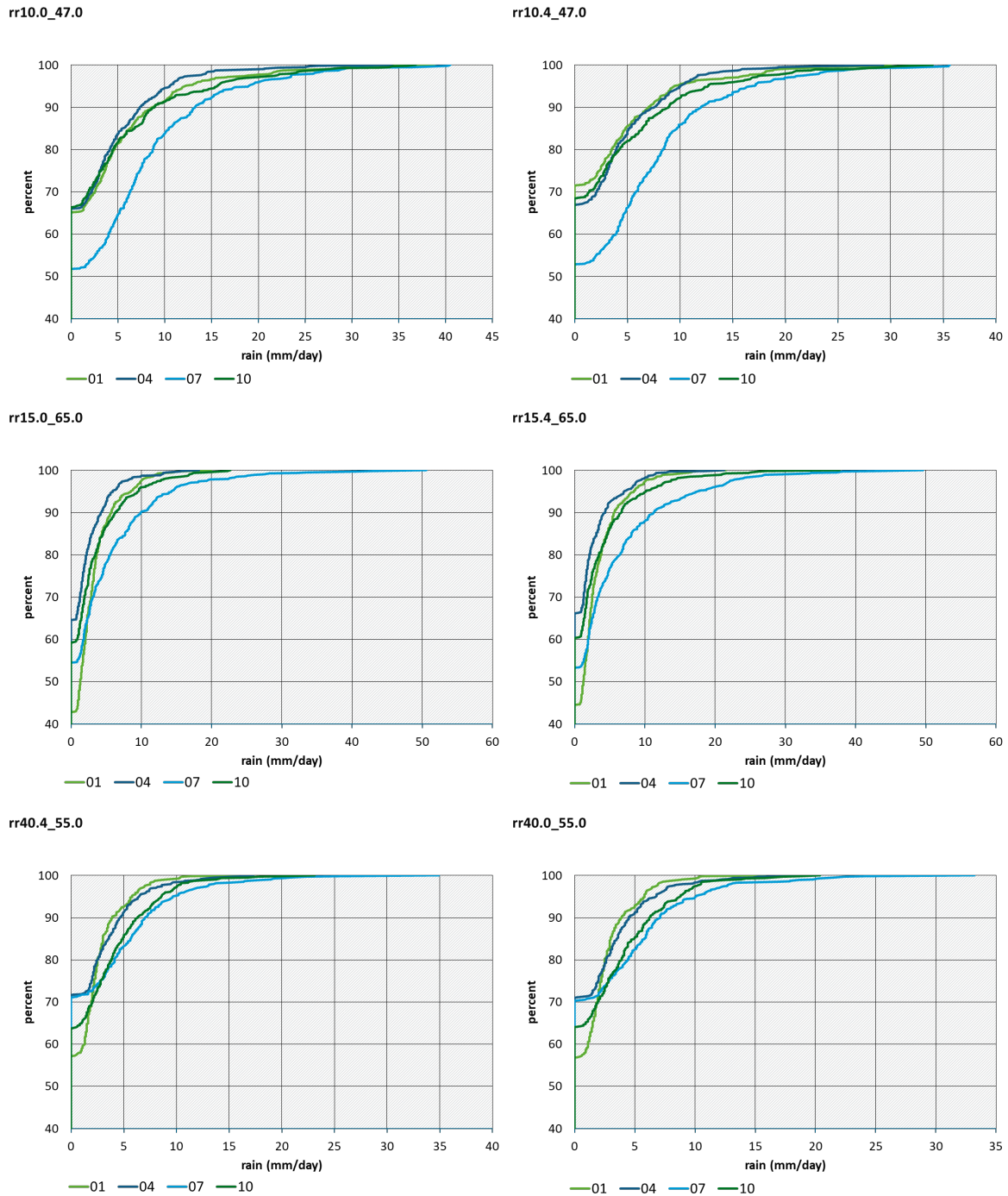
**tg10.0_47.0**
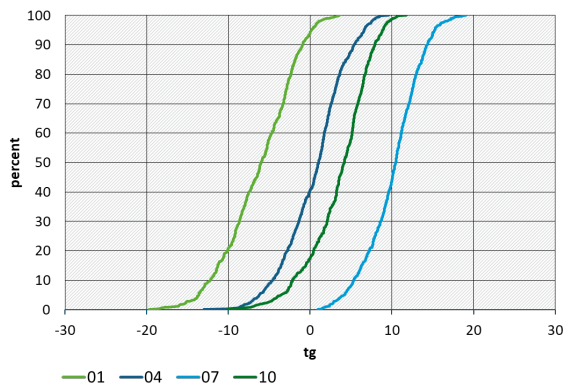


**tg10.4_47.0**
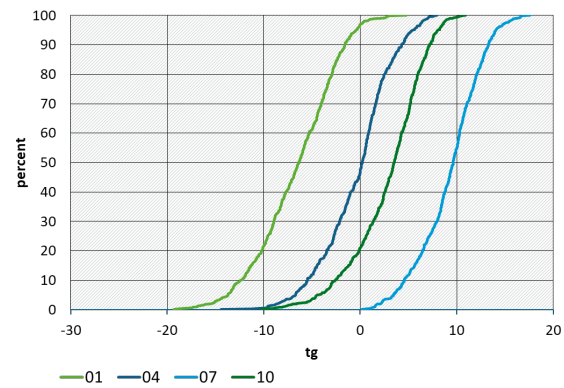


**tg15.0_65.0**
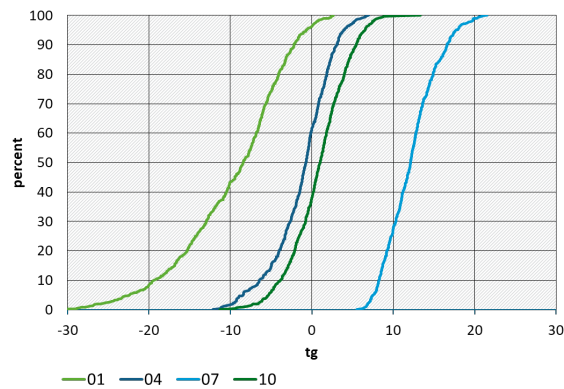


**tg15.4_65.0**
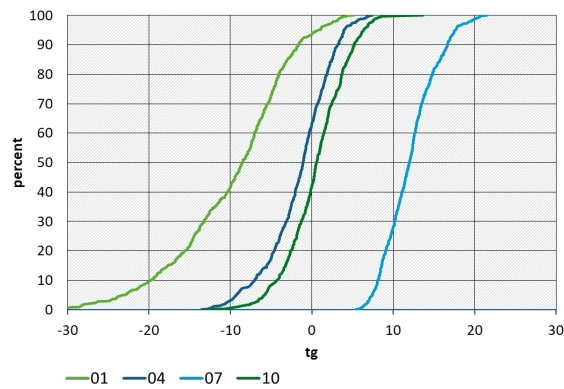


**tg40.0_55.0**
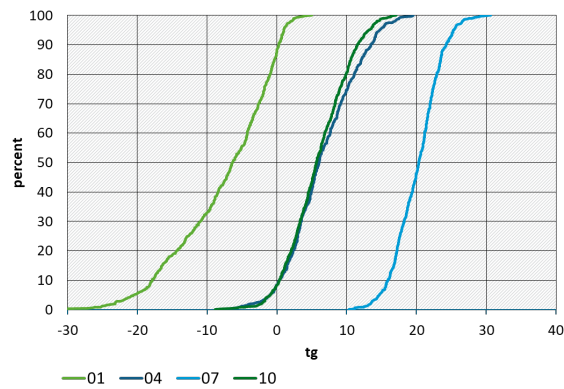


**tg40.4_55.0**
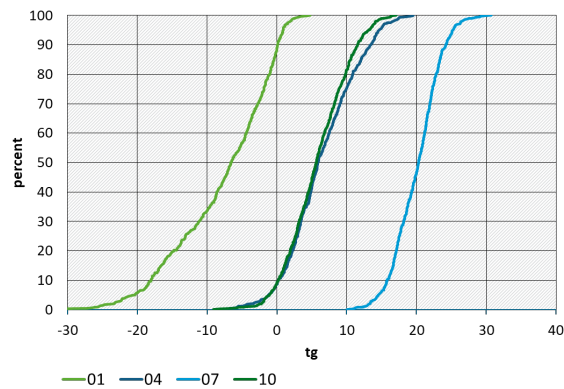


Source: Own representation, WUR,IIASA,RIVM

### 4.3.2      Computing critical loads for Europe: MainLoop.R

MainLoop.R is the principal script for computing critical loads. It reads all data needed, prepares the meteorological data for MetHyd by interpolating missing values, runs the MetHyd model and runs the critical load model. Details are provided in Chapter 5.

## 4.4    Verification of the results

To verify that the R scripts give the same results as the original Fortran codes, the following test was made:

► The R script was run for the Netherlands, computing the critical loads and also writing out the basic input data: site location, soil type, landuse type, soil texture class, soil weathering rate class, distance to coast, forest growth region, temperature and leaching flux.

► The original Fortran program was run with these inputs to compute critical loads:  the above-mentioned input data were used to compute, for example, nutrient uptake, weathering rates, base cation deposition, and finally critical loads.

► Results from both runs were compared to see if the R procedure yields the same results as the original Fortran procedure.

Note that this is a technical verification only of the software. The comparison of the new critical loads with the critical loads published in the latest RIVM-CCE report is provided in Chapter 6.

# 5  Computing critical loads using the R software

## 5.1  Setting up the R environment

The software to compute critical loads was written in R. In order to make use of it, the following steps have to be taken:

(1) Install R 3.6.3 and R studio 1.3.959 as well as Rtools (rtools40-x86_64.exe)

(2) Unpack the zip-archive with the datafiles and the R software to a proper root directory ; for example, we have used e:\data\projects\CL_CCEUBA. In this report this directory will be referred to as <ROOTDIR>. The zipped files comprise R codes, R projects and data files (for example soil data, forest growth data etc.)

(3) Run R studio with administrator rights. Install the following packages from the CRAN repository:
   a.  raster
   b.  mgcv
   c.  foreach
   d.  rworldmap
   e.  rworldxtra
   f.  rgdal
   g.  devtools
   h.  lubridate
   i.  matrix

   Make sure they are installed on the R program directory, not somewhere on a network drive!

(4) Set the working directory to the root of the software <ROOTDIR> !!

(5) Run the script <ROOTDIR>\BGRun\scripts\**MKeps.R**. This stores a globally used value for the accuracy of some computations. The 'eps' value is one very small floating point value (1.e-7). By storing it in a data structure (eps.rda) it can be assessed by different scripts, and does not need to be defined more than once.

(6) Install the package BGDB which contains the basic routines for critical load computations; on the R command line use:

```
devtools::install_local('<ROOTDIR>/BGDB/BGDB',repos = NULL,
type="source")
```

This will generate a few warnings that can be ignored. The BGDB package will be created.

(7) Download the meteo data for future use. This is a one-time action. This only needs to be repeated if the E-OBS /CRU data sets are updated/improved

   a.  Register for downloading the E-OBS meteorological data at:

   https://surfobs.climate.copernicus.eu/dataaccess/access_eobs.php

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Click on *register as an E-OBS user*

After registration, download the 20.e versions of the data files at https://surfobs.climate.copernicus.eu/dataaccess/access_eobs.php#datafiles;

Download the following six files:

Version 20.0e, 0.1° regular grid, ensemble mean, the files for variables

TG, TN, TX, RR, PP and QQ.

The data files are in netcdf format and occupy together about 15 GB. Store these files in the directory

      <ROOTDIR>\PreData\METncs

b. Acquire the global meteorological data from the Climatic Research Unit

    https://crudata.uea.ac.uk/cru/data/hrg/cru_ts_4.04/cruts.2004151855.v4.04/

for the variables:

cld, pre, tmp and wet

as the gzipped netcdf files (nc). Unzip and store these in the following directory :

      <ROOTDIR>\PreData\METglobs

(8) Make sure (the ArcGIS script should take care of it) that the files with the map overlay results are in <ROOTDIR>/BGRun/SoilData, and each has its own directory with the files in it. The name of this directory must be equal to the ISO 3-letter country code

(9) Setup is now complete. After preparing the meteorological data (see next section), one can run the critical load script

## 5.2 Preparing the meteorological data

In R studio load the BGRUN.proj. ***Set the working directory to the project location***!!

Run the script **cropMetNC.R**. This will read the netcdf files with EObs data and create RDS files with meteo data in <ROOTDIR>BGRun\PreData\METcrops. There will be one file for each 0.1° latitude stripe, 460 files in total. This process will take considerable time (in the order of 22 hours on a Desktop PC with Windows 10 and 8 Gb memory) to complete. The files will contain the daily meteorological data for the latitude stripe for the period 1998–2018 (i.e. 20 years of data plus one year for initialization (1998)).

To prepare the global meteorological data for use in EUDB 2020, run the script **MkCrudeMET.R**. This script processes the netcdf files with global data and produces a file with an R data frame called CrudeMET.RDS in the directory <ROOTDIR>BGRun\PreData.

## 5.3    Running the critical load model: MainLoop.R

In R studio load the BGRUN.proj. ***Set the working directory to the project location!***

Open the script **MainLoop.R**. This is the script that computes the critical loads by setting parameters, reading in data and computing input data.

Then the script can be run. If CalcMeteo and CalcHyd are set to true (first run), running critical loads for the full geographical extent will take considerable computing time (several days on a Windows 10 desktop with 8 Gb of memory). Later runs (with meteorological data and MetHyd results available) go very fast, and take about ten minutes on the same desktop.

MainLoop.R is the principal script for computing critical loads. It reads all data needed, prepares the meteorological data for MetHyd by interpolating missing values, runs the MetHyd model and runs the critical load model. The following steps are carried out by MainLoop and some inputs by the user are required:

(1)    Set criteria for the critical loads: iAci and critAci. iAci is the criterion to be used for computing the maximum critical load for S (CLmaxS) (Table 9):

**Table 9: iAci values and their associated criterion**

| iAci | Criterion | iAci | Criterion |
|------|-----------|------|-----------|
| 1 | molar [Al]:[Bc] | 5 | 5: critAci = [ANC] (eq/m$^3$) |
| 2 | [Al] (eq/m$^3$) | 6 | 6: critAci = molar [Bc]:[H] ([Al]=0 => not compatible with VSD!) |
| 3 | EBc (base saturation) (fraction, NOT%) | 7 | 7: critAci = Alw ! no Al depletion |
| 4 | pH | 8 | 8: critAci = molar [Al]:[Ca] |
|  |  | 11 | 11: critAci = [Al]:[Bc], only if [Al]>0.1 |

And critAci is the critical value for the chosen criterion (e.g. 1 for Al/Bc = 1 mol/mol).

(2)    Specify whether meteorology must be prepared and/or MetHyd must be run, by setting CalcMeteo and CalcHyd to T or F (True or False). For the first run for a (set of) countries these values MUST be set to T(rue). For later runs, the values can be set to F(alse) if there is no need to re-do the hydrology (i.e. if meteorological data are unchanged and the MetHyd model is unchanged). The preparation of meteorological data and computation of hydrology with MetHyd take considerable time, so if MetHyd has been run once, both values can be set to F to considerably speed up the critical load calculations.

(3)    Basic settings are made in **EUCLpar.R** on, e.g., soil depth and the wood density of trees. **constSettings.R** defines which ions are included in the Ions data type (a data type used to store e.g. values for concentrations, weathering rates and uptake).

(4)    **Get_Inp.R** defines all other model parameters such as aluminium dissolution constant and the dissociation constant for organic acids.

(5)    Growth and base cation (BC) deposition are read from files. Growth is read from two files _Efiscen_2020.csv for the areas where Efiscen results are available, _RegGrow.tab for the other regions. The latter files gives the growth rates directly, growth rates for Efiscen regions are computed in **MkTreeGrowth.R** by dividing the total increment of a species in the region (in 1000m$^3$) by the area of this species in the Efiscen region (in 1000 ha); the _Efiscen_2000.csv contains standard Efiscen output and can thus be replaced by

results from future Efiscen data. The script **PrepBCdeps.R** reads the BC deposition for that part of the EMEP region for which these data are available (file: BCTotAve.dep). For the rest of Europe and Eurasia, the total Ca deposition is read from the file TotalCaS.asc, which is used to compute all other base cation deposition as well (see section 2.6).

(6)     Countries for which the model(s) need to be run are defined by their ISO-3166 2-letter code by setting the *Countries2Prep* variable. The countries for which critical loads can be computed are:

AL AT BA BE BG BY CH CZ DE DK EE ES FI FR GB GR HR HU IE IT LI LT LU LV MD ME MK MT NL NO PL PT RO RS RU SE SI SK UA

(7)     Read the soil map information: each polygon is a Soil Mapping Unit (SMU) that consists of a number of Soil Typological Units (STUs; the actual soil types) each with a relative area within the SMU. This information is available in the file _Scode_1M_EAw.Tab. For each STU, this file contains also information on soil texture, water regime and weathering rate class.

(8)     Read the file _SoilChar.Tab that contains data for pH, organic carbon and nitrogen and CaCO3 content per soil type based on ICP Forest Level I monitoring data. For critical loads, only the pH is used from this set to estimate DOC (dissolved organic carbon) concentrations.

(9)     Determine the clay, sand and silt content of the soil based on the STU's texture class.

(10)    Read the meteorological data previously prepared with **MkCrudeMet.R**.

(11)    Get all rasters (.tif files) from the overlay procedures (**GetSoilMap.R**) and then select those rasters that belong to the countries that have been selected (**ReadSoil.R**); these files contain the information on soil, land use, forest growth region, N2k region and distance to coast for 0.01 × 0.01 degree cells.

(12)    Start the master loop over the stripes of 0.5 degrees. First determine if meteorology needs processing and/or MetHyd need to be run and if so, carry out these tasks (**MetHyd.R**) . If not, read the stored results from a previous MetHyd run from the proper .rds data file.

(13)    Get from the already read-in data the base cation deposition for this latitude stripe by storing it for the relevant array of EMEP cells.

(14)    Compute the weathering rates for all soils in the latitude stripe.

(15)    Compute the uptake of nutrients for each receptor in the latitude stripe by (a) setting the proper forest type (conifers, deciduous, mixed) based on the landuse from the map overlay tif file; if the landuse is a non-forest (e.g. grasslands or heather) net uptake is set to zero; (b) find the area of these forest types within the region from the Efiscen results; (c) compute the forest area-weighted mean nutrient contents; and (d) compute the average uptake for each cell by multiplying the average nutrient contents by the average area-weighted forest growth. For non-Efiscen regions, nutrient contents for conifers, deciduous and mixed forests are used without further weighing.

(16)    For critical loads based on base saturation (iAci = 3), set the exchange model to Gapon and set the exchange constants to standard values.

(17)    Run the SMB model.

(18)    Make a dataframe (*AllData*) to store the results from SMB and the relevant input data for this latitude stripe. For each latitude stripe, store these dataframes in a list of frames (*sLAices*).

(19)    After all latitude stripes have been processed, write out the list of dataframes with input data and SMB results to an ASCII-file (EUBGSMB.csv) in the *BGRUN/output* directory.

Results are written to the file EUBGSMB.csv  in <ROOTDIR>\BGRUN\output. The name of the file can be adjusted in the script for subsequent runs. It contains the 'standard' critical load data base output (Table 10):

**Table 10: Parameters in the output file of the critical load procedure (MainLoop); all in eq/ha/yr unless said otherwise**

| Item | Description | Item | description |
|------|-------------|------|-------------|
| Lon | Longitude of the site (°) | Nawe | Na weathering |
| Lat | Latitude of the site (°) | Caupt | Ca uptake |
| EcoArea | Area of the receptor (km$^2$) | Mgupt | Mg uptake |
| Protection | N2k area indicator (if applicable) | Kupt | K uptake |
| EUNIS | EUNIS class | Nupt | N uptake |
| crittype | Chemical criterion used | Nimacc | N immobilization (long term) |
| critvalue | Value of the Chemical criterion | Qle | Leaching flux (precipitation surplus; m/yr) |
| cNacc | Critical N concentration (eq/m$^3$) | fde | Denitrification fraction (-) |
| CLmaxS | Maximum critical load of S | lgKalox | Log  of equilibrium constant for Al(OH)$_3$ dissolution (mol/L)$^{-2}$ |
| CLminN | Minimum critical load of N | expAl | Exponent in Al(OH)$_3$ dissolution equation (-) |
| CLmaxN | Maximum critical load of N | cOrgAcids | Concentration of organic acids (mol/m$^3$) |
| CLnutN | Critical load of nutrient N | temp | Temperature (°C) |
| thick | Thickness of the rootzone (m) | cnt | Country (ISO 2-letter code) |
| nANCcrit | ANC leaching | i50 | EMEP 50 I index |
| Cadep | Ca deposition | j50 | EMEP50 J index |
| Mgdep | Mg deposition | Soil | FAO Soil type |
| Kdep | K deposition | itex | Texture class |
| Nadep | Na deposition | ipm | Parent material class |

| Item | Description | Item | description |
|------|-------------|------|-------------|
| Cawe | Ca weathering | Iftype | Forest type |
| Mgwe | Mg weathering | EFISCEN | Efiscen region |
| Kwe | K weathering | | |

## 5.4    Verification of the R procedures

A comparison with the results from the 2017 critical loads based on Fortran routines (Posch & Reinds, 2017) has been made (see section 4.4) by aggregating the results from EUDB 2020 runs to combinations of EMEP cell, soil type, soil texture, water regime, forest type and Efiscen region for the Netherlands (Figure 4). The 2017 runs with the Fortran software have been repeated using the new forest growth and precipitation surplus data, so that we can verify that the software yields identical results. The comparison shows that all inputs to the critical load equations are identical (weathering, uptake, leaching) as well as the computed CLminN. For CLmaxS a minor deviation was found, due to some rounding issues in the input data of the gibbsite equilibrium constant.

**Figure 4: Verification of the R procedures**



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**CLmaxS**
**EUDB 2020 versus CCE 2017**



**CLminN**
**EUDB 2020 versus CCE 2017**
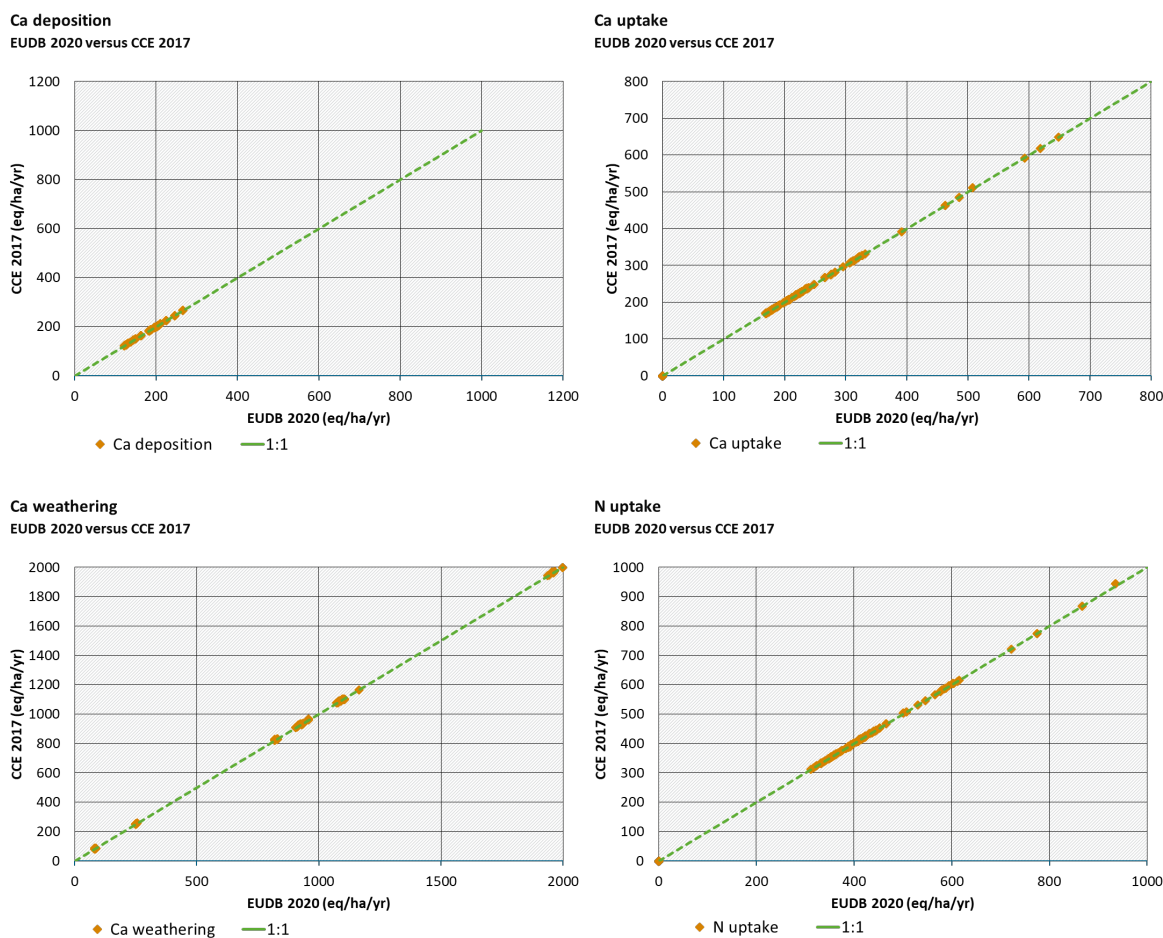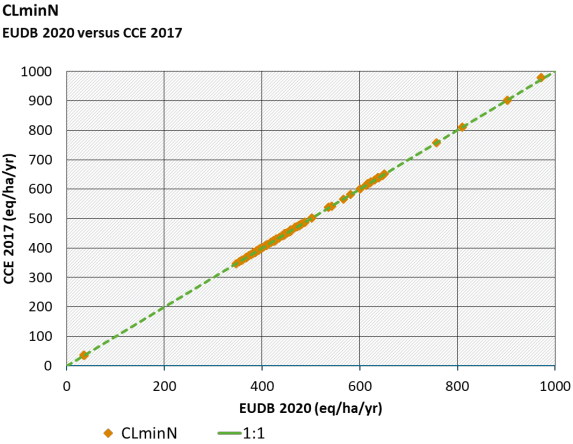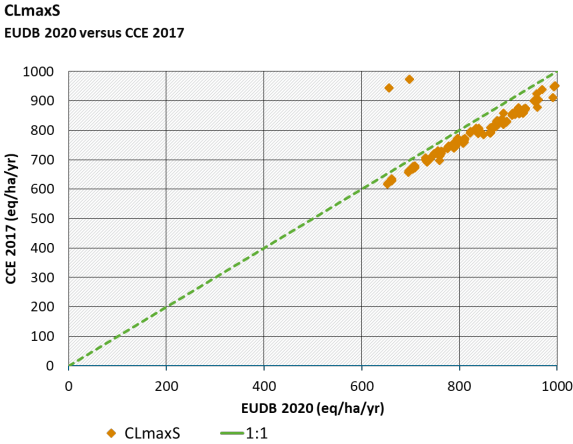


Source: Own representation, WUR,IIASA,RIVM

# 6   Comparison with 2017 background data base results

This chapter shows the results from a full run with EUDB 2020. For comparison, results are also shown from the background critical loads computed by RIVM-CCE and reported in the CCE Final Report 2017 (Posch & Reinds, 2017).  Maps of some input data as well as maps of CLmaxS, CLmaxN and CLnutN per 0.5°×0.25° grid cell are provided, using the median value of input variables such as deposition and weathering rates and of critical loads. For the critical loads also the 5th percentile maps are shown. For CLminN, cumulative distribution functions (CDFs) are given. Although differences with the existing critical load database may arise from the update of the underlying data sets (see below), patterns and magnitude of critical loads should be comparable to earlier results, also to avoid issues in future use of the new critical loads in integrated assessments.

Compared to the 2017 results a few changes have been made regarding the computation of critical loads:

1. The software was ported to R
2. The MetHyd model uses daily data for 1999-2018 instead of monthly data 1970-2000
3. The Efiscen forest growth data have been updated to the latest (2016) version (Petz et al., 2016; Prins et al., 2017)

In the maps shown below, for Natura2000 areas all receptors are included, outside Natura2000 only receptors > 0.2 km[2] are used to be consistent with how the 2017 results were calculated. An overview of the area and total number of receptors per country and EUNIS class is given in Appendix A4.

**Figure 5: Median Calcium deposition (Cadep) in eq/ha/yr for the 2017 background DB (left) and the EUDB 2020 (right)**
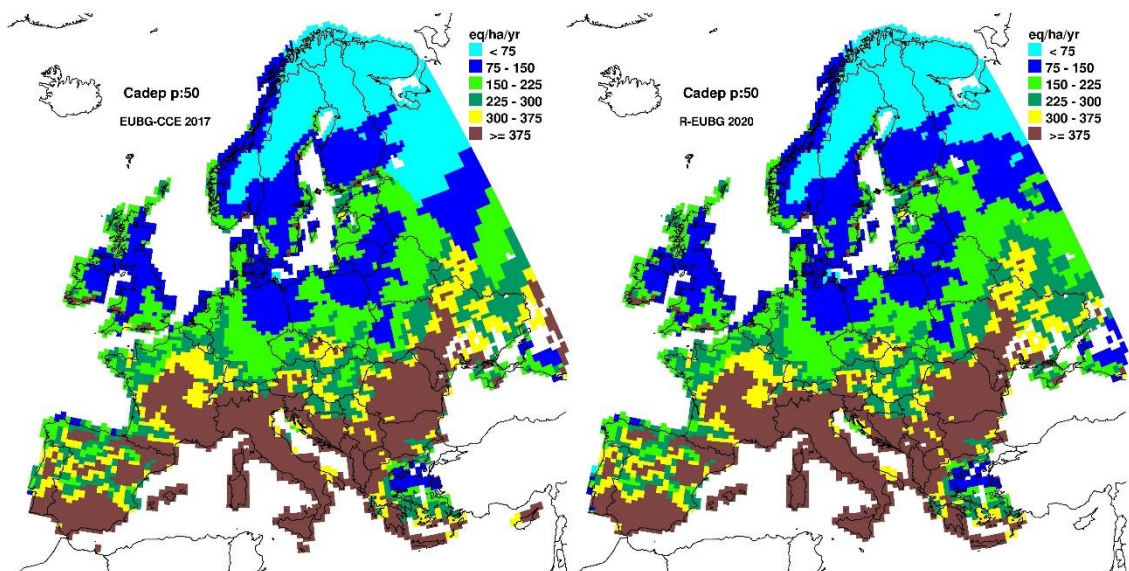


Source: Own representation, WUR,IIASA,RIVM

Calcium deposition patterns from the R procedure are identical to the patterns in the 2017 data for most of Europe (Figure 5)
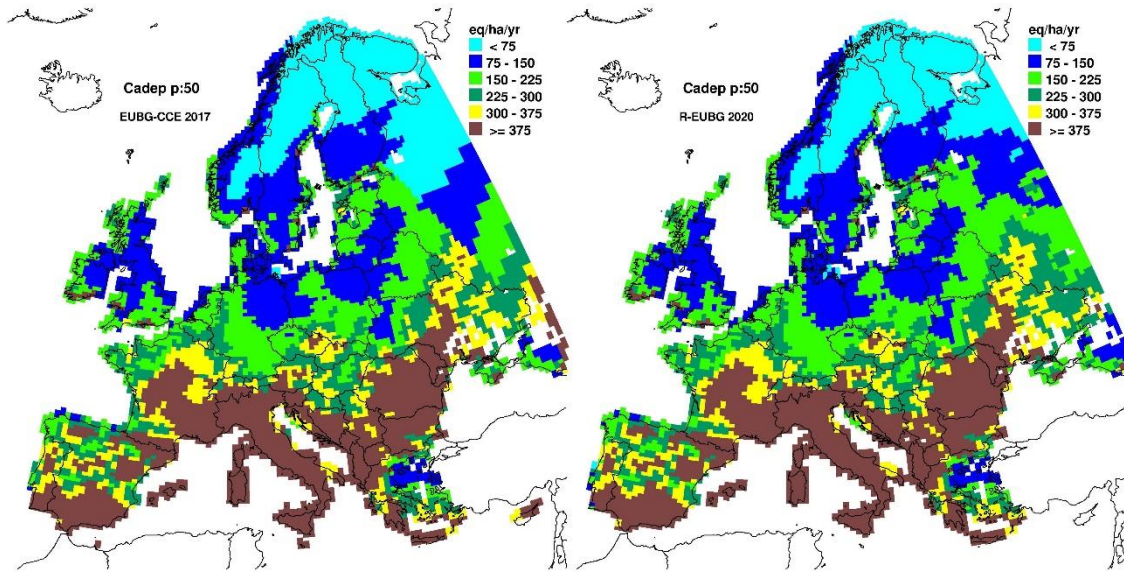
(as they should be). Only in the north-western part of Russia, differences occur, but the new results show a smoother transition with neighbouring countries than the 2017 results. It seems that in the 2017 calculations there may have been a problem with the Ca deposition (interpolation) estimates in this area.

**Figure 6: Median Magnesium deposition (Mgdep) in eq/ha/yr for the 2017 background DB (left) and the EUDB 2020 (right)**
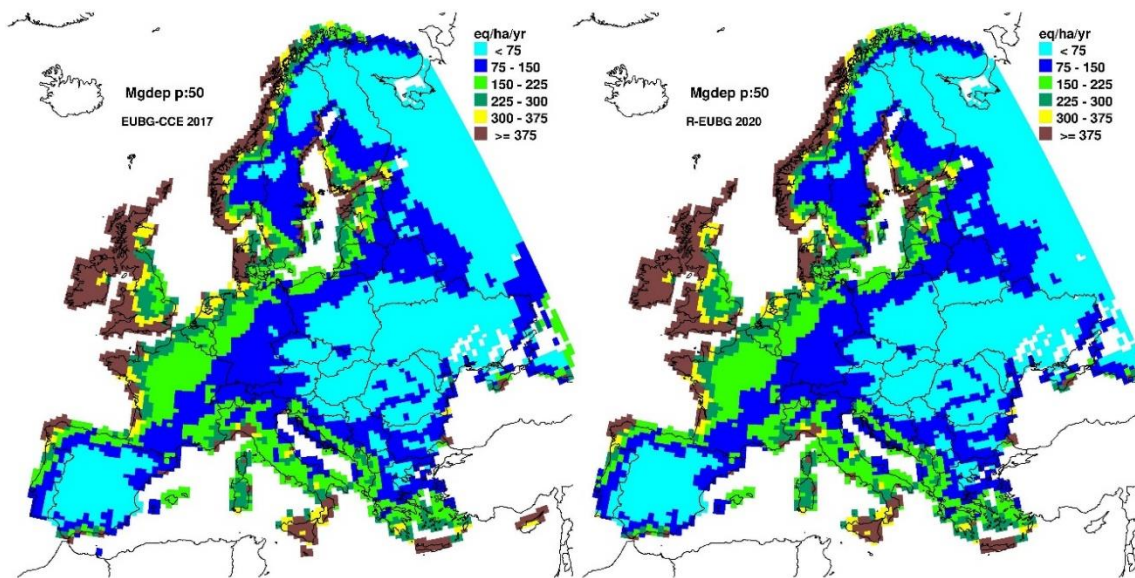


Source: Own representation, WUR,IIASA,RIVM

Magnesium deposition (Mgdep) patterns from the R procedure are also almost identical to the patterns in the 2017 data for most of Europe (Figure 6). Substantial part of the Mgdep is from seasalt origin, as the deposition decreases with the distance to the coast.

**Figure 7: Median Calcium weathering (Cawe) in eq/ha/yr for the 2017 background DB (left) and the EUDB 2020 (right)**
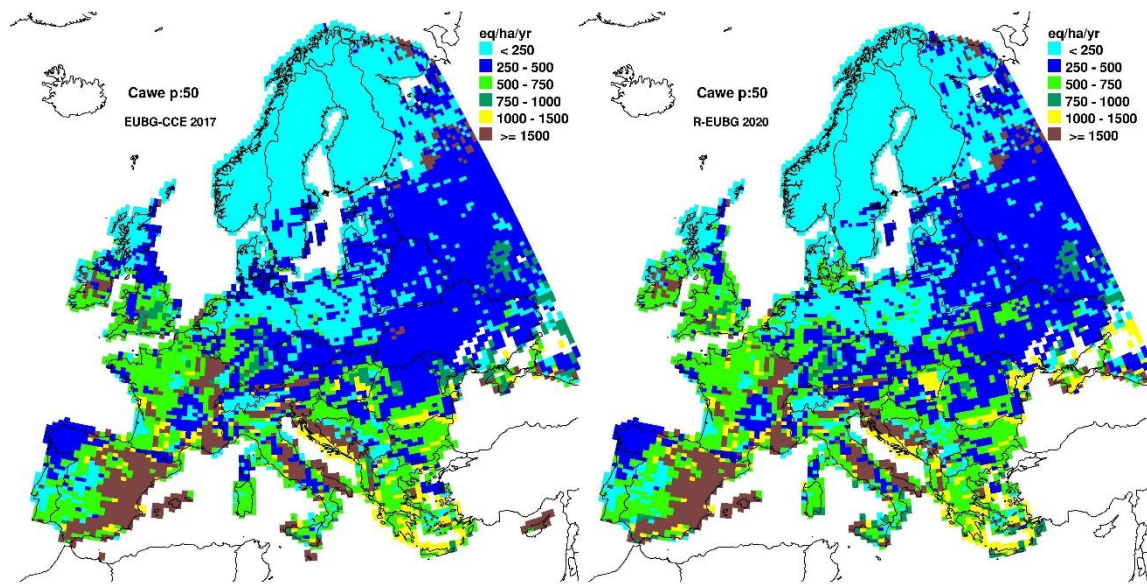


Source: Own representation, WUR,IIASA,RIVM

The patterns in the weathering rates for Calcium (Ca) compare well (Figure 7) between EUDB 2020 and the 2017 results. Since the weathering rate not only depends on soil type, parent material and soil texture, but also on temperature, small changes have occurred due to the use of a different set of meteorological data (see section 4.3.1 for details).

**Figure 8: Median precipitation surplus (leaching flux; Qle) in mm/yr for the 2017 background DB (left) and the EUDB 2020 (right)**
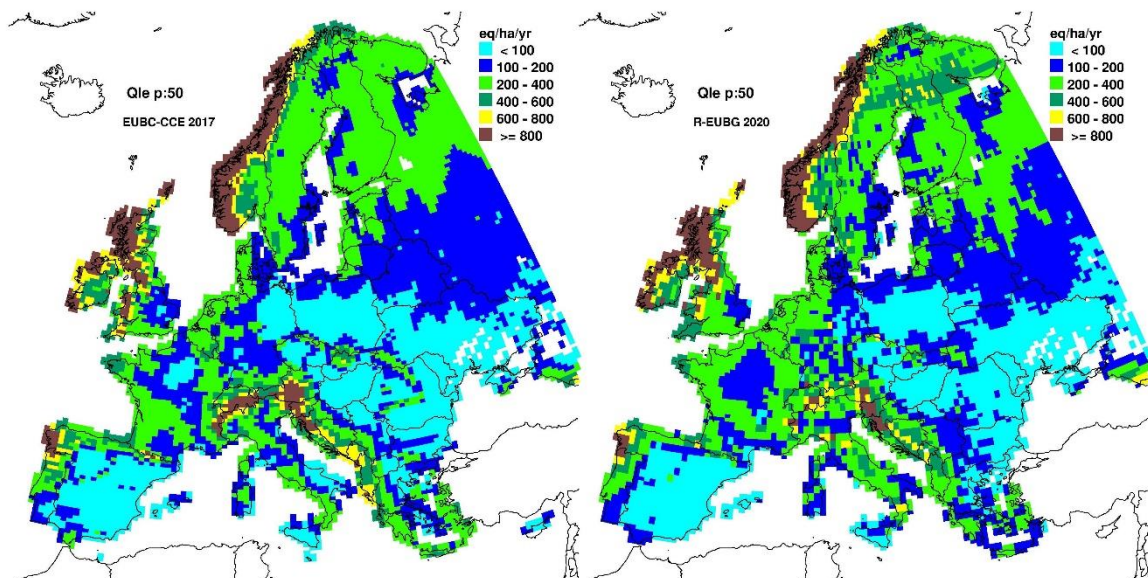


Source: Own representation, WUR,IIASA,RIVM

The precipitation surplus patterns over Europe show strong similarities in the two data bases: areas with high precipitation surpluses are found in mountainous areas, Scotland and the western part of Norway, low precipitation surpluses in Spain and central Europe (Figure 8). The

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simulated precipitation surplus in parts of the Alpine region are now somewhat lower than in 2017, either because the precipitation in this area is lower than in the older data set or because increased temperatures lead to higher evapotranspiration.

**Figure 9: Cumulative distribution function (CDF) of CLminN in eq/ha/yr for the 2017 background DB (green line) and EUDB 2020 (blue line)**
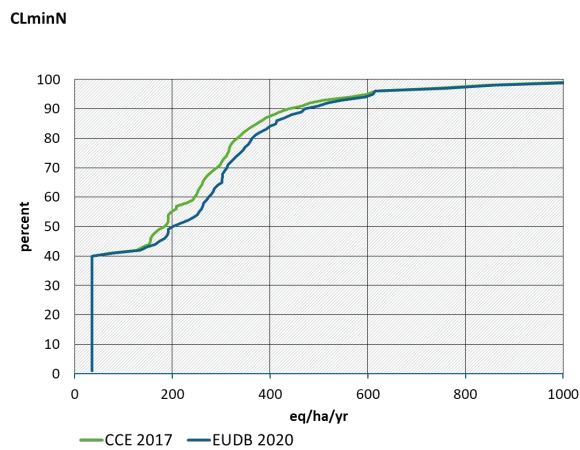


CLminN

Source: Own representation, WUR,IIASA,RIVM

The minimum critical load of N, CLminN, is very similar in both simulations (Figure 9). Differences stem from the use of the new Efiscen forest data as CLminN is mostly determined by N uptake. Maps for ClminN are not shown as the 5 percentile maps would, in almost all cells, display the same value of CLminN for non-forest (i.e. the value of $N_{imm}$).

**Figure 10: 5-th percentile (upper row) and median (lower row) CLmaxS in eq/ha/yr for the 2017 background DB (left) and the EUDB 2020 (right)**
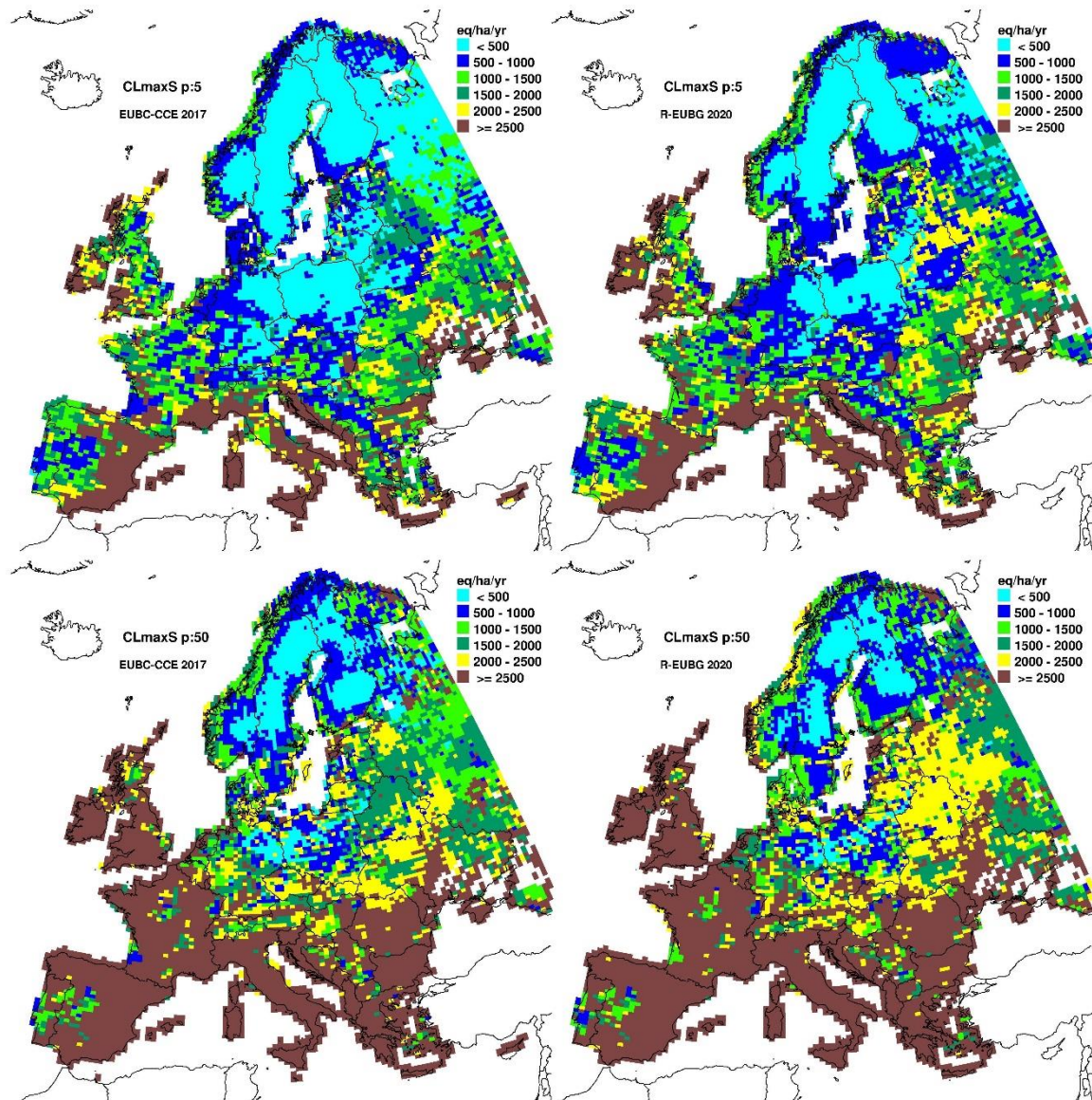


Source: Own representation, WUR,IIASA,RIVM

The geographical patterns for the maximum critical load for S, CLmaxS, show strong similarities (Figure 10). Some differences occur in Russia due to the change in base cation deposition. Minor differences can occur because of minor changes in uptake (due to updated growth data) and/or because leaching has changed (due to the use of a different meteorological data set; see Figure 8).

The geographical patterns for the critical load for eutrophying N, CLnutN, also show strong similarities (Figure 11). CLnutN consist of N immobilisation (set to a constant value, in our case 1 kgN/ha/yr), net N uptake (zero for non-forests), the N denitrification fraction and N leaching. Lowest values (see the 5 percentile maps) are confined to non-forest ecosystems as the net uptake term is zero for them. Since the N immobilisation is constant, the 5 percentile maps reflect the patterns in precipitation surplus in Europe (see Figure 8).

**Figure 11: 5-th percentile (upper row) and median (lower row) CLnutN in eq/ha/yr for the 2017 background DB (left) and the EUDB 2020 (right)**
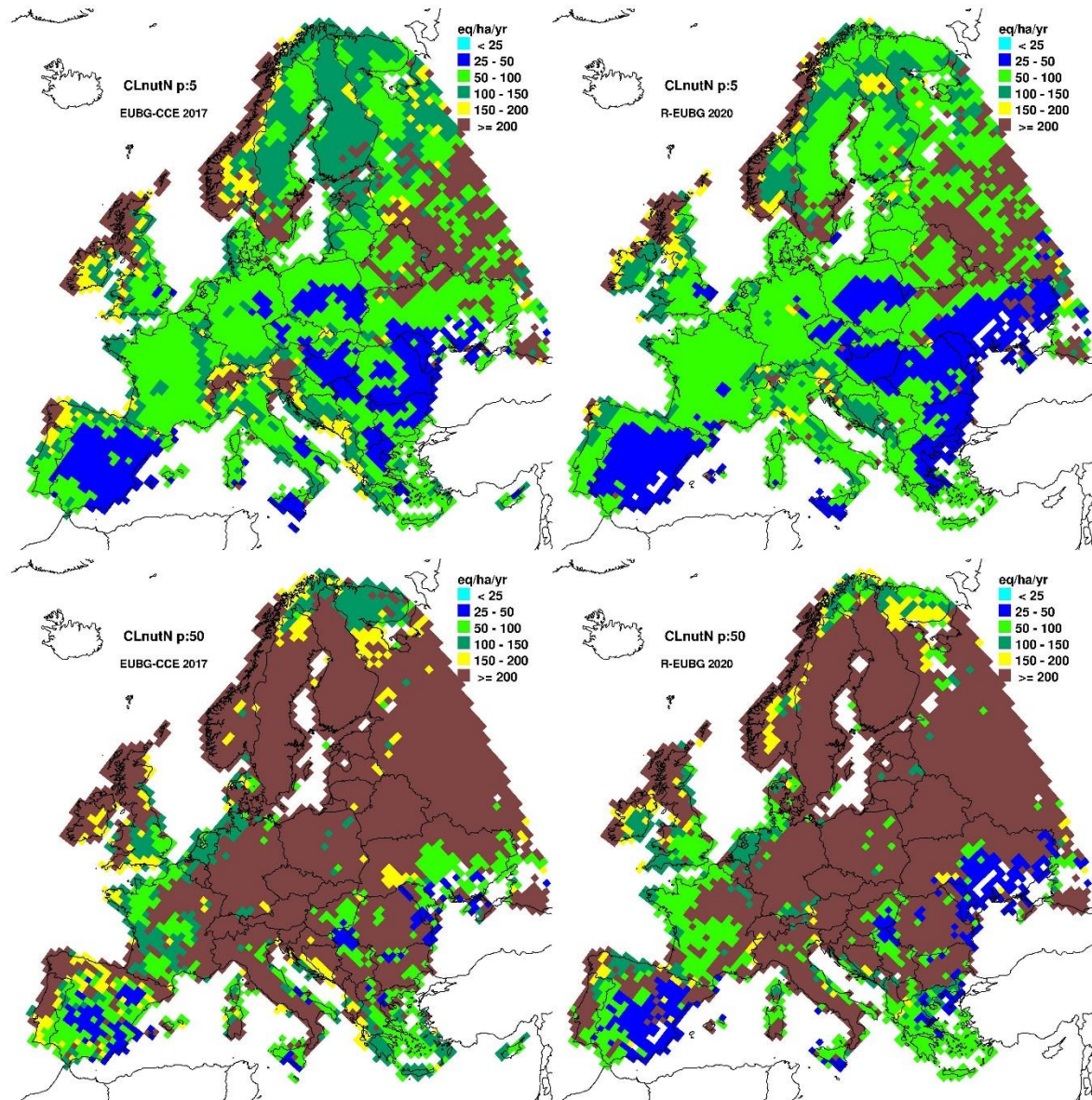


Source: Own representation, WUR,IIASA,RIVM

# 7   Comparison of critical loads with national data from Ireland and Germany

## 7.1   Introduction

In this chapter the CL data bases provided in 2017 by the Irish and German NFCs are compared with the CLs for those countries generated by the new R-software for producing the 2020 European Background Dababase (EUDB 2020). We focus on a comparison of ecosystem areas and of the critical loads of N and S. To explain some of the differences, also some individual terms in the critical loads are shown. The equations used to compute the critical loads are provided in Chapter 3.

## 7.2   Ireland

In the number of sites and ecosystem areas in Ireland for the Level-1 EUNIS classes in the 2017 NFC data and the new EUDB 2020 are compared. The following points spring to the eye:

► The Irish NFC provided almost double the number of sites on less than a third of the area compared to the EUDB 2020, which probably indicates that the national data are based on more detailed maps of soil, vegetation etc, leading to more (unique) sites.

► The Irish NFC provided no acidity CL data for EUNIS class D ('mires, bogs and fens').

► Concerning forests (EUNIS class G), Ireland provided more than ten times the number of sites than that in the EUDB 2020, covering almost double the area. This could be due to a different definition of a 'forest'.

 the number of sites and ecosystem areas in Ireland for the Level-1 EUNIS classes in the 2017 NFC data[1] and the new EUDB 2020 are compared. The following points spring to the eye:

► The Irish NFC provided almost double the number of sites on less than a third of the area compared to the EUDB 2020, which probably indicates that the national data are based on more detailed maps of soil, vegetation etc, leading to more (unique) sites.

► The Irish NFC provided no acidity CL data for EUNIS class D ('mires, bogs and fens').

► Concerning forests (EUNIS class G), Ireland provided more than ten times the number of sites than that in the EUDB 2020, covering almost double the area. This could be due to a different definition of a 'forest'.

**Table 11: Number of sites and ecosystem areas in Ireland for the Level-1 EUNIS classes in the 2017 NFC data and the new EUDB 2020. Note that Ireland provided data for 193824 terrestrial sites covering an area of 18345.1 km$^2$ (for 153000 sites both CLaci and CLeutN were given, covering 12652.4 km$^2$).**

| EUNIS | Number of sites (records) | | | | Ecosystem area (km$^2$) | | | |
|---|---|---|---|---|---|---|---|---|
| Code | NFC | | | EUDB | NFC | | | EUDB |
| | CLaci | CLeutN | Both | Both | CLaci | CLeutN | Both | Both |
| A | 0 | 223 | 0 | 0 | 0 | 15,9 | 0 | 0 |

| EUNIS | Number of sites (records) | | | | Ecosystem area (km²) | | | |
|---|---|---|---|---|---|---|---|---|
| Code | NFC | | | EUDB | NFC | | | EUDB |
| | CLaci | CLeutN | Both | Both | CLaci | CLeutN | Both | Both |
| D | 0 | 33295 | 0 | 21412 | 0 | 5373,8 | 0 | 11351,5 |
| E | 63011 | 63203 | 62460 | 67429 | 6971,7 | 7017,1 | 6946,2 | 43814,9 |
| F | 2660 | 2714 | 2660 | 1186 | 351,2 | 354,5 | 351,2 | 572,5 |
| G | 87880 | 93838 | 87880 | 9079 | 5355 | 5558,2 | 5355,0 | 2916,8 |
| SUM | 153551 | 193273 | 153000 | 99106 | 12677,9 | 18319,6 | 12652,4 | 58655,7 |

Table 11 lists the FAO soil types found in Ireland and the ecosystem area they cover (in EUDB 2020); these include calcareous soils as well (Ic, Ec, Gc) which will be associated with high values of CLmaxS.

**Table 11: FAO soil types in Ireland and ecosystem area which they cover in EUDB 2020**

| Soil type | Ecosystem area (km²) | Soil type | Ecosystem area (km²) |
|---|---|---|---|
| Bd | 4163.3 | Gh | 2181.3 |
| Bds | 2813.3 | I | 1444.8 |
| Be | 4877.8 | Ic | 52.6 |
| Bec | 420.9 | Id | 2554.2 |
| Bg | 110.9 | Je | 82.9 |
| De | 181.6 | Lg | 362.9 |
| Ec | 150.3 | Lo | 8755 |
| Eo | 52.6 | Od | 7650.7 |
| Gc | 1369.6 | Odp | 1666.1 |
| Gd | 2083.2 | Oe | 1229.8 |
| Gds | 6375.4 | Ph | 6.3 |
| Ge | 2663.3 | Po | 2675.5 |
| Ges | 174.4 | Pp | 4556.7 |

The national critical loads for Ireland have been computed with:

► A critical pH of 4.2

► Base cations merged into Ca, and Mg deposition assumed to be from sea salt origin completely

► The gibbssite equilibrium constant is a function of soil type (lgKAlox = 6.5 for peat soils, lgKAlox =7.5 for lithosols, regosols and leptic podzols and lgKAlox =8 for all other soils)

For comparison and to show the influence of the above listed 'deviations' from a standard run with Al/Bc =1 as the criterium, CLs for Ireland in the new EUDB 2020 have been computed for 3 different options ('Runs') which are analysed here:

► **Run1**: Standard run with Al/Bc = 1, all BC ions separate, fixed lgKAlox (8.7 for mineral soils, 6.7 for peat)

► **Run2**: Run with critical pH = 4.2 and BC merged in Ca, Mg deposition considered as sea-salt, fixed lgKAlox (8.7 for mineral soils, 6.7 for peat)

► **Run3**: Run with critical pH = 4.2 and BC merged in Ca, Mg deposition considered as sea-salt and lgKAlox as a function of soil type

Figure 12 compares CLmaxS as computed by the NFC and for the new EUDB 2020 by plotting their respective CDFs; in particular for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph). This shows that Run1 leads to much higher critical loads for S than when the national approach is used, due to the high ANC leaching fluxes when using Al/Bc=1.

For Run1 ANC leaching (eq/ha/yr) varies between 622.5 and 18809.2, for Run2 between 129.3 and 2083.8, and for the NFC data between -3155.9, -95.6. Run3, which mimics the national approach best, shows comparable CLmaxS values especially for forests, although it seems that the national results from Ireland do not include calcareous soils, which the EUDB 2020 results do, leading to a 'jump' in the CDF caused by an area with high critical loads.

**Figure 12:** CDFs of CLmaxS for forests (EUNIS 'G'; left) and non-forests (all other classes) in Ireland taken from the 2017 NFC data (green lines) and the 3 Runs of the new EUDB 2020 (blue lines; see text for their definition)
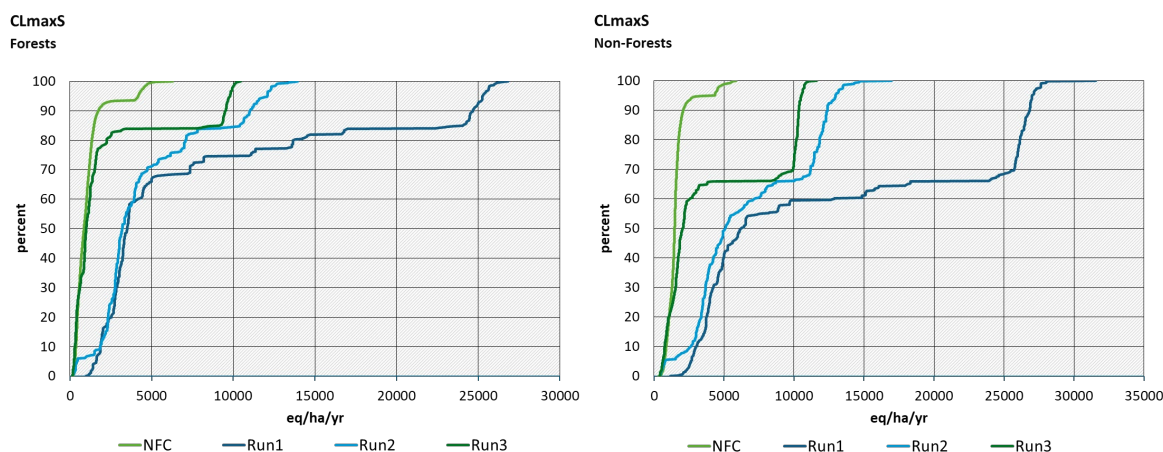


Source: Own representation, WUR, IIASA,RIVM, Irish NFC

Figure 13 compares CLminN as computed by the NFC and for the new EUDB 2020 by plotting their respective CDFs; in particular for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph).

**Figure 13: CDFs of CLminN for forests (EUNIS 'G'; left) and non-forests (all other classes) taken in Ireland from the 2017 NFC data (green lines) and the new EUDB 2020 (blue lines)**
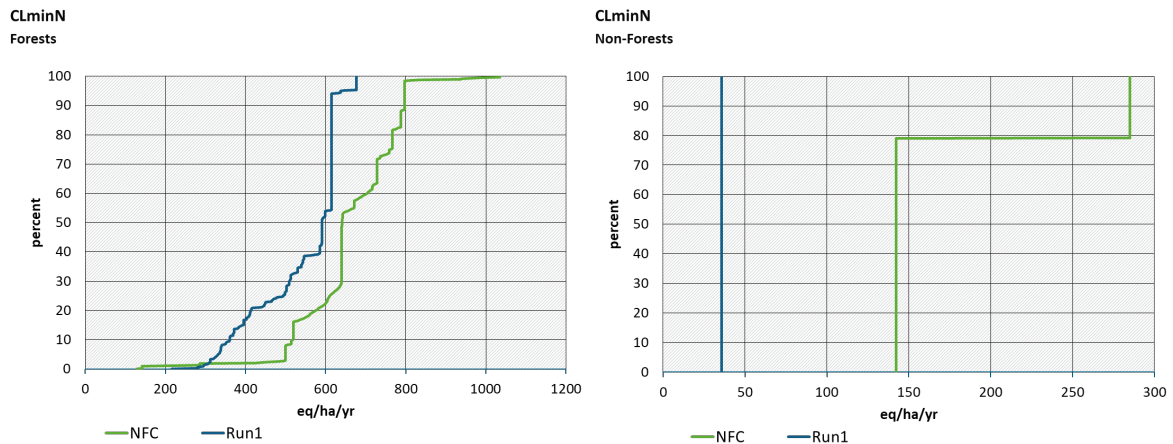


Source: Own representation, WUR, IIASA, RIVM, Irish NFC

Obviously, this quantity is the same for all 3 Runs; and the differences depend (only) on the resp. assumptions/choices of the net uptake (for forests) and immobilization of N; interesting is the jump in NFC CLminN for non-forests; for some ecosystems either the N immobilisation is different or some net N uptake has been included (in EUDB 2020 the net uptake for non-forest ecosystems is zero and CLminN consists of N immobilisation only).

Figure 14 compares CLmaxN as computed by the NFC and for the new EUDB 2020 by plotting their respective CDFs for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph). The CDFs reflect the peculiarities of the CDFs of CLmaxS (Figure 12), and to a minor extent those of CLminN (Figure 13), and in addition the 'choice' of the N denitrification flux (see equation 3.3). Also here, Run3 gives the best fit between the national data and the EUDB 2020 results.

**Figure 14: CDFs of CLmaxN for forests (EUNIS 'G'; left) and non-forests (all other classes) in Ireland taken from the 2017 NFC data (green lines) and the 3 Runs of the new EUDB 2020 (blue lines; see text for their definition)**



Source: Own representation, WUR, IIASA, RIVM, Irish NFC

Figure 15 compares CLeutN as computed by the NFC and CLnutN from the new EUDB 2020 by plotting their respective CDFs for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph).

**Figure 15: CDFs of CLnutN/CLeutN for forests (EUNIS 'G'; left) and non-forests (all other classes) in Ireland taken from the 2017 NFC data (green lines) and the new EUDB 2020 (blue lines)**
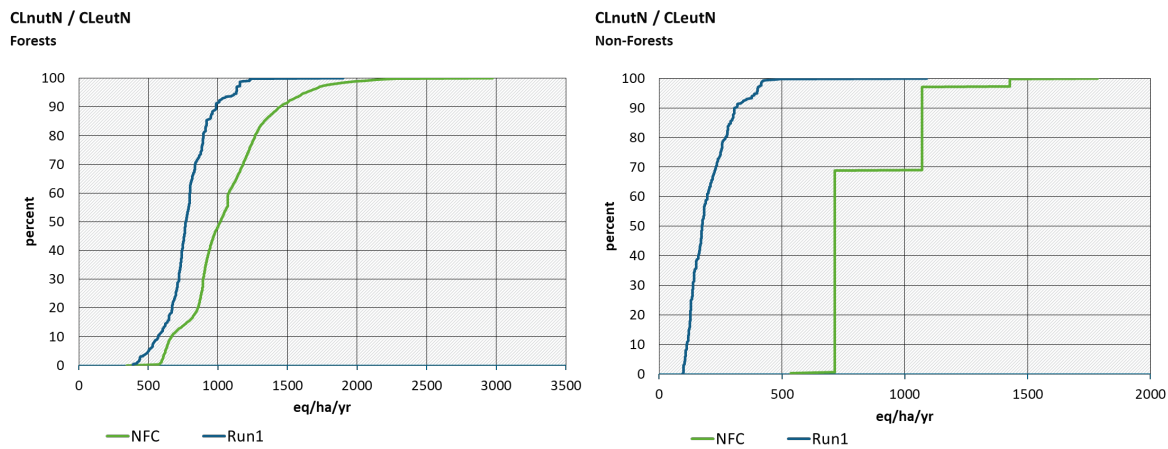


Source: Own representation, WUR,IIASA,RIVM, Irish NFC

Obviously, this quantity is the same for all 3 Runs; and the differences depend (only) on the differences in CLminN (see above) and the chosen critical limit (+ runoff). Differences can also be caused by the fact that for some ecosystems Ireland may have used only empirical CLs for N, leading to higher CLeutN.

## 7.3    Germany

For Germany only one run was made with EUDB 2020, using Al/Bc =1 as the critical limit. The national data from Germany have been computed using various criteria (pH, Bsat, Al/Bc, no Al depletion) and various critical values for each of these criteria (e.g. critical base saturation between 3% and 62% and critical pH between 4.08 and 6.2) in many classes (Schlutow et al., 2017). Since the assignment of criteria and their values was based on national maps of soils and vegetation, there is no possibility to mimic this in EUDB 2020.

In Table 12 the number of sites and ecosystem areas in Germany for the Level-1 EUNIS classes in the 2017 NFC data and the new EUDB 2020 are compared. The following points spring to the eye:

► The German NFC provided more than double the number of sites on roughly the same area compared to the EUDB 2020, most likely because the maps on soil, vegetation, etc. used by the German NFC are much more detailed than the maps on the European scale of the EUDB 2020, leading to a higher number of (unique) receptors.

► However, about one third of the NFC's ecosystem area are marine habitats (EUNIS class A) – this is 'compensated' by a much smaller (1/40-th) area for EUNIS class E (grasslands). This may be (partly) caused by a difference between the EUNIS map used in EUDB 2020 and the national vegetation map of Germany used by the NFC.

**Table 12: Number of sites and ecosystem areas in Germany for the Level-1 EUNIS classes in the 2017 NFC data and the new EUDB 2020**

| EUNIS | | Number of sites (records) | | Ecosystem area (km²) | |
|---|---|---|---|---|---|
| Code | description | NFC | EUDB 2020 | NFC | EUDB 2020 |
| A | Marine habitats | 1464 | 0 | 50826.5 | 0 |
| D | Mires, bogs and fens | 15596 | 5147 | 830 | 1319.7 |
| E | Grasslands and lands dominated by forbs, mosses and lichens | 36939 | 169735 | 1391.2 | 55563.2 |
| F | Heathland, scrub and tundra | 10106 | 2285 | 803.7 | 625.4 |
| G | Woodland, forest and other wooded land | 1202042 | 303124 | 99347.4 | 103941.1 |
| **SUM** | | 1266147 | 480291 | 153198.9 | 161449.4 |

Figure 16 compares CLmaxS as computed by the NFC and for the new EUDB 2020 by plotting their respective CDFs; in particular for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph). While for forests the CDFs are close for about 90% of the area, for non-forests the NFC data are pretty close to zero for most sites whereas EUDB 2020 critical loads are much higher. This is probably due to the strict way the German national critical loads have been computed: for each receptor the minimum value of the critical loads based on different criteria is used; if for one of the criteria the CL becomes very low for non-forest ecosystems, the resulting critical loads will be very low as well.

**Figure 16: CDFs of CLmaxS for forests (EUNIS 'G'; left) and non-forests (all other classes) in Germany taken from the 2017 NFC data (green lines) and the new EUDB 2020 (blue lines)**
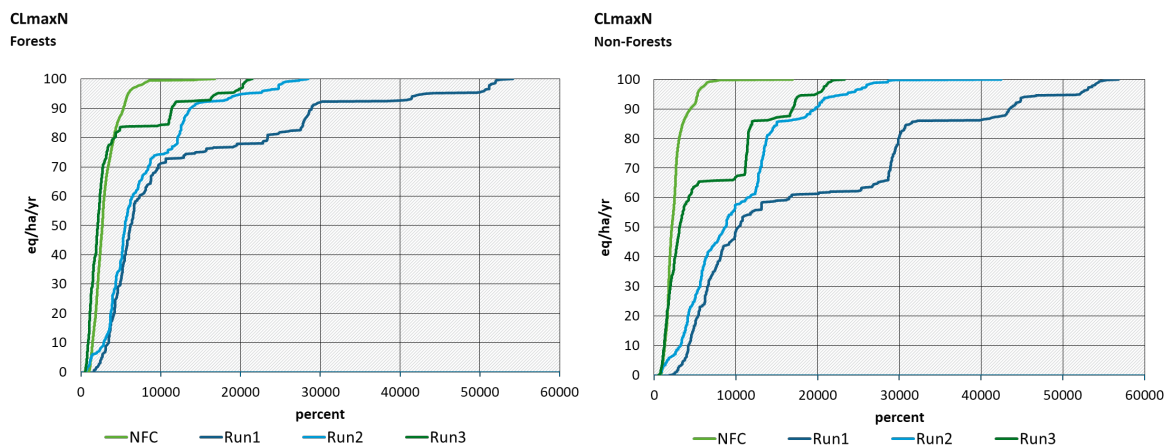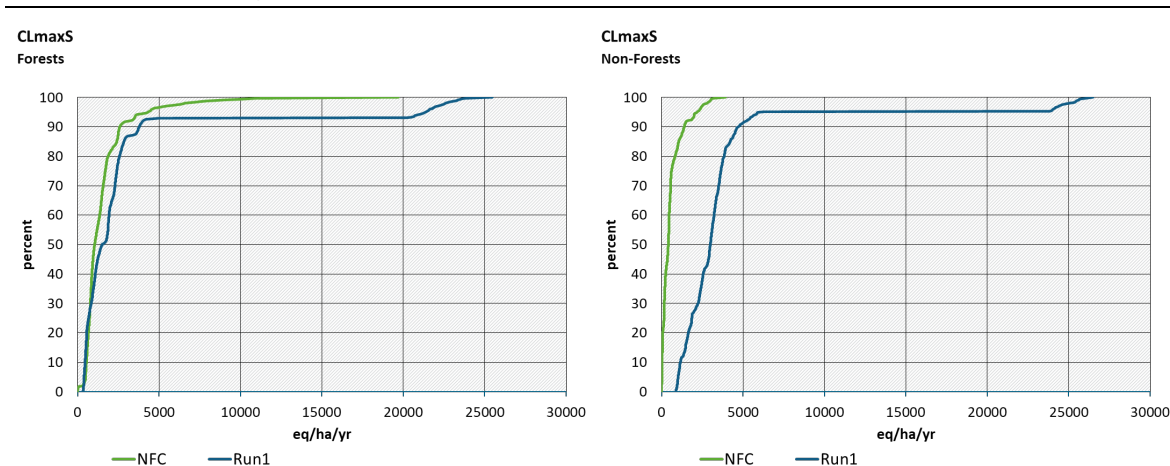


Source: Own representation, WUR,IIASA,RIVM, German NFC

Figure 17 compares CLminN as computed by the NFC and for the new EUDB 2020 by plotting their respective CDFs; in particular for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph). Obviously, the differences depend (only) on the resp. assumptions/choices of the net uptake (for forests) and immobilization of N.

**Figure 17: CDFs of CLminN for forests (EUNIS 'G'; left) and non-forests (all other classes) in Germany taken from the 2017 NFC data (green lines) and the new EUDB 2020 (blue lines)**
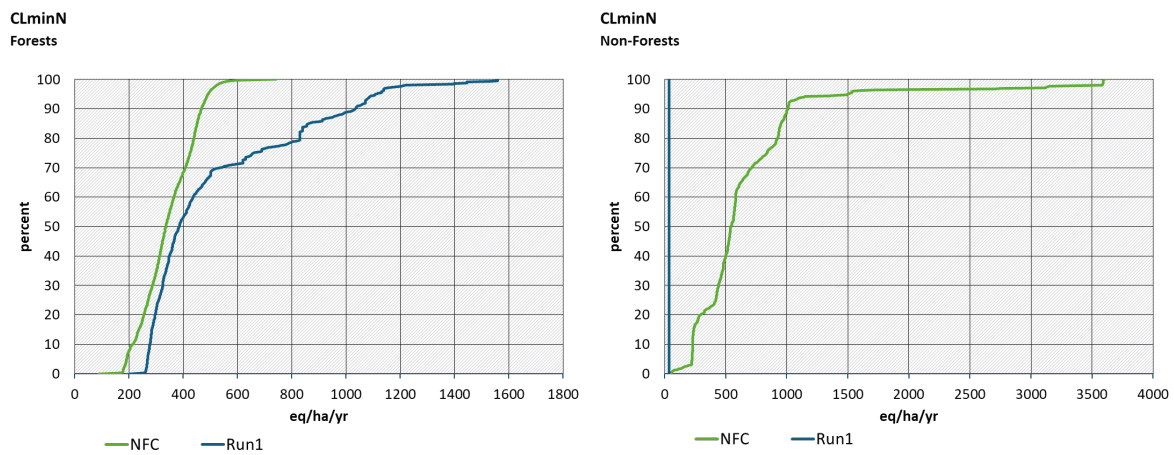


Source: Own representation, WUR,IIASA,RIVM, German NFC

The CLminN values for non-forest in the NFC data are much higher than when using EUDB 2020 because for EUDB for non-forests we assume that they are unmanaged so there is no harvest to remove N, and CLminN therefore consists of N immobilisation only (see Equation 3.2). The German NFC however assumes net removal of N also from these systems.

This difference is mainly caused by differences in net N uptake (Figure 18). The national CLminN is based on net N removal rates in non-forest ecosystems up to 1200 eq/ha/yr. N immobilisation is somewhat higher than in EUDB 2020, but the difference is limited. Germany uses a different approach for computing $N_{imm}$ than the constant value used in EUDB 2020 (see (Schlutow et al., 2017).

**Figure 18: CDFs of long-term net N immobilisation ($N_{imm}$, top) and net N uptake ($N_{upt}$, bottom) for forests (EUNIS 'G'; left) and non-forests (all other classes) in Germany taken from the 2017 NFC data (green lines) and the new EUDB 2020 (blue lines)**

Source: Own representation, WUR,IIASA,RIVM, German NFC

Figure 19 compares CLmaxN as computed by the NFC and for the new EUDB 2020 by plotting their respective CDFs for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph). The CDFs reflect the peculiarities of the CDFs of CLmaxS (Figure 16), and to a minor extent those of CLminN (Figure 17), and in addition the 'choice' of the denitrification fraction $f_{de}$.

**Figure 19: CDFs of CLmaxN for forests (EUNIS 'G'; left) and non-forests (all other classes) in Germany taken from the 2017 NFC data (green lines) and the new EUDB 2020 (blue lines)**



Source: Own representation, WUR,IIASA,RIVM, German NFC

Figure 20 compares CLeutN as computed by the NFC and CLnutN from the new EUDB 2020 by plotting their respective CDFs for forests (EUNIS 'G'; left graph) and all other ecosystems (right graph). Obviously, the differences depend (only) on the differences in CLminN (see above) and the chosen critical limit (+ runoff). Differences are larger for non-forest ecosystems than for forest ecosystems: maybe for some ecosystems Germany has used only empirical CLs for N, leading to higher CLeutN.

**Figure 20: CDFs of CLnutN/CLeutN for forests (EUNIS 'G'; left) and non-forests (all other classes) in Germany taken from the 2017 NFC data (green lines) and the new EUDB 2020 (blue lines)**
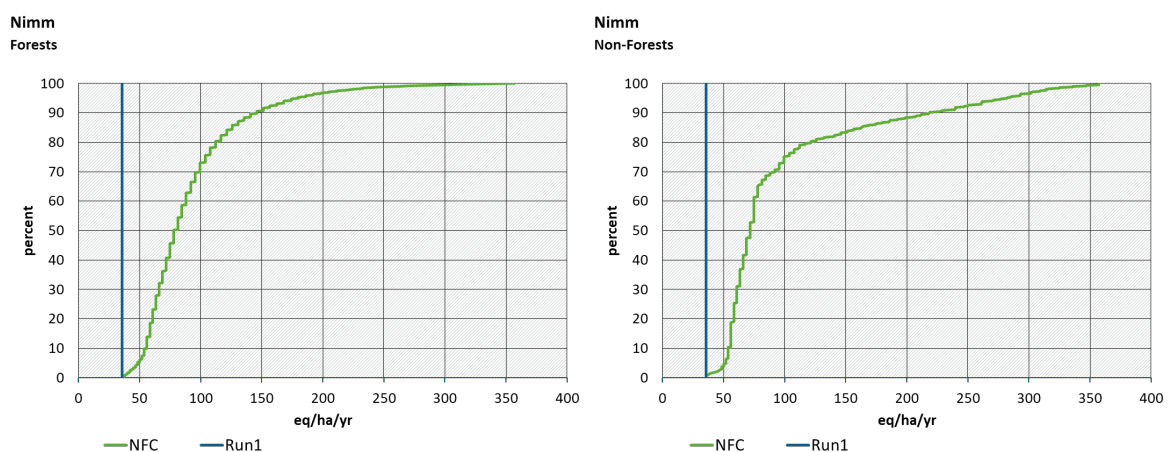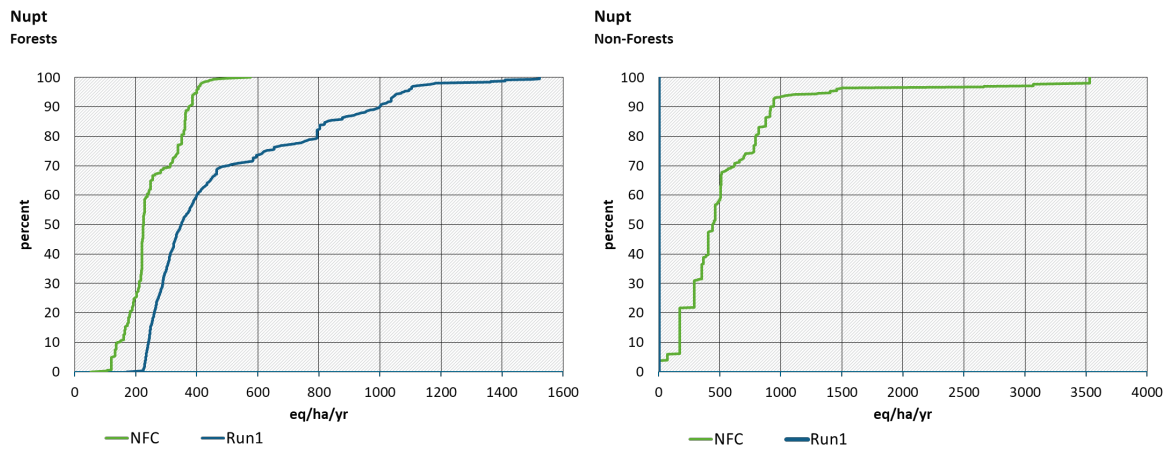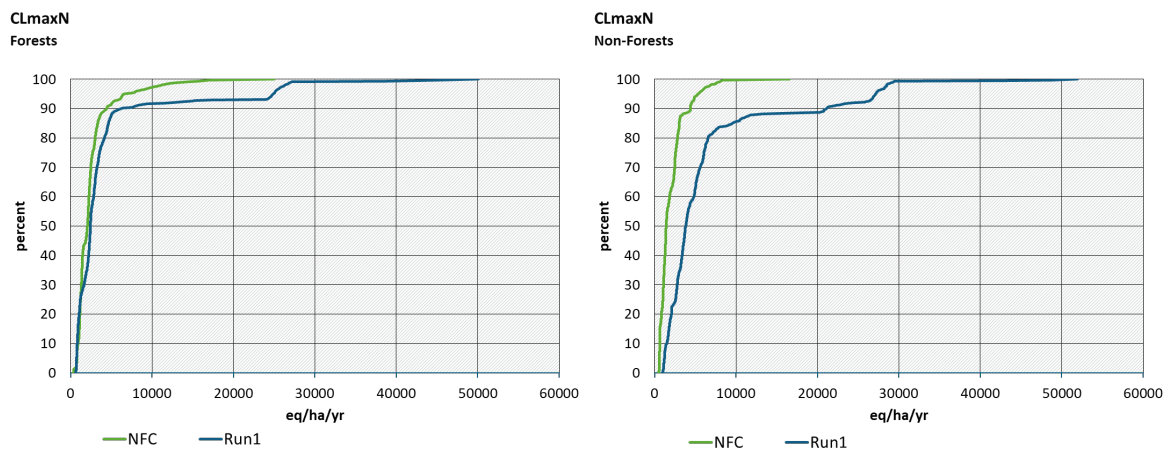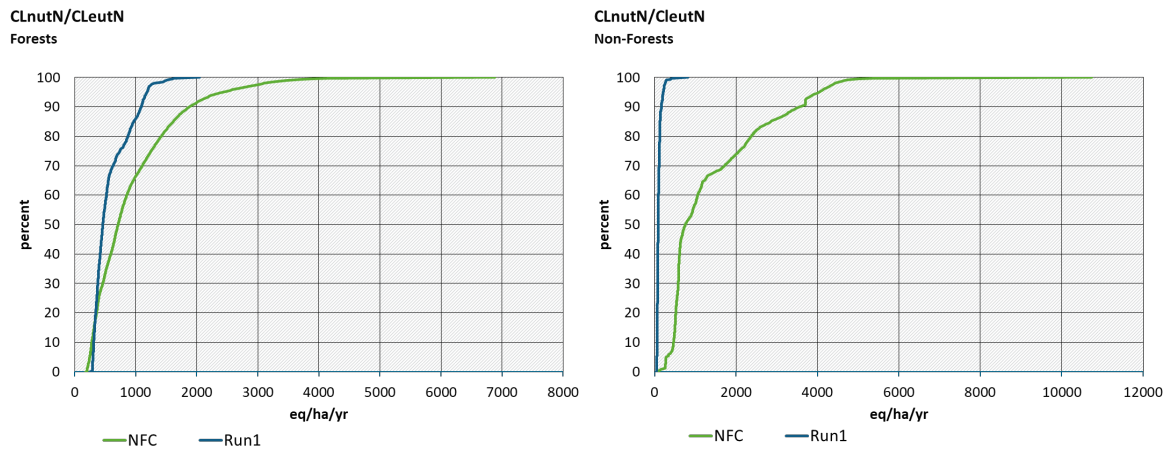


Source: Own representation, WUR,IIASA,RIVM, German NFC

# 8 Comparison of computed critical loads of eutrophication (or nutrient N) to empirical critical loads

## 8.1 Introduction

The critical load of eutrophication (or nutrient N) in SMB is computed according to equation 3.1:

$$CL_{nut}N = N_i + N_u + \frac{Q \cdot [N]_{acc}}{1 - f_{de}}$$

where $N_i$ is the long-term net immobilisation of N in the soil, $N_u$ is the net removal of N in harvested vegetation, $f_{de}$ ($0 \leq f_{de} < 1$) is the fraction of the net N input denitrified, $Q$ is the precipitation surplus (runoff) leaving the soil compartment (rooting zone), and $[N]_{acc}$ is the acceptable (critical) N concentration to avoid 'harmful effects' on the chosen 'sensitive element of the environment'. This critical N concentration is often set to 0.2 mg N.l⁻¹ for conifers forest and to 0.3 mg N.l⁻¹ for deciduous forests and seminatural vegetations according to the Mapping Manual. De Vries et al. (2015) argue that these values are representative for vegetation changes in Northern Europe and values for western European ecosystems are probably higher.

Empirical critical N loads are mostly based on observed changes in the structure and functioning of ecosystems in field studies, and relate to unwanted changes in species abundance, composition and/or diversity ('ecosystem structure'), or to N leaching, -decomposition or mineralisation rate ('ecosystem functioning') (e.g. Bobbink and Hettelingh (2011)).

From the above two definitions, it is clear that although both critical loads mainly relate to the eutrophying effects of N, they are conceptually different. The SMB critical load has a strong leaching component: for non-forests for example, net uptake is set to zero, acceptable N immobilisation is a low, constant, value for all EUNIS classes, so N leaching is the main term in equation 3.1. For empirical critical loads, the mentioned changes in species abundance, composition and/or diversity may be caused by N enrichment in the soil organic and mineral phase without resulting in enhanced N leaching.

## 8.2 Method

For all receptors CLnutN has been computed using equation 3.1. In the standard run values for the critical N concentration of 0.2 mg N.l⁻¹ for conifers forest and 0.3 mg N.l⁻¹ for deciduous forests and seminatural vegetations were used. A second run was made with values of 3 mg N.l⁻¹ for conifers and deciduous forests and to 3.5 mg N.l⁻¹ for seminatural vegetations; the latter run uses critical values thought to be representative for vegetation changes in Western Europe (see De Vries et al. (2015)). Next, ecosystem-area weighted percentiles have been computed for the 30 available terrestrial EUNIS classes. These values have been compared to empirical critical loads. It should be noted that the empirical critical loads are defined as ranges (e.g. 5-10 kg N/ha/yr). In the tables we show both this range as well as the mean value (i.e. the mean of the minimum and maximum value of the range).

## 8.3 Results

When using the strictest values for the critical N concentration, SMB based CLnutN varies mostly between 1-3 and 10-15 kg N.ha⁻¹ (Table 13). Compared to empirical critical loads, the median value is mostly still lower than the lower end of the empirical range. If we assume an average precipitation surplus of 300 mm/yr and a critical concentration of 0.3 mg.l⁻¹, the leaching flux will be about 1 kg of N.ha⁻¹. For non-forests, net N uptake is zero and N immobilisation is set to

0.5 kgN .ha$^{-1}$ for all ecosystems, so CLnutN for non-forest ecosystems will be 1.5 kg.ha$^{-1}$ on average, which is indeed about the median SMB-based value found for the non-forest ecosystem in Table 13. Ecosystems that also occur in areas with a very high precipitations surplus (and thus a much higher N leaching), such as for example 'Temperate shrub heathlands', can have very high critical loads, as expressed by the 95 percentile values. CLnutN values for forests are higher than for non-forest ecosystems because net uptake for forests always has a positive value.

**Table 13: SMB critical loads (5, 50 and 95 percentile) using the [*N*]$_{acc}$ criterium for northern Europe and empirical critical loads (lower value of the range, upper value and mean) in kg N.ha$^{-1}$**

| EUNIS | 5 | 50 | 95 | Empirical | | | Description |
| --- | --- | --- | --- | --- | --- | --- | --- |
| | | | | min | max | avg | |
| 4100 | 1 | 2 | 7 | 5 | 10 | 7.5 | Raised and blanket bogs |
| 5120 | 1 | 1 | 1 | 15 | 25 | 20 | Perennial grasslands and basic steppes |
| 5179 | 1 | 1 | 10 | 8 | 15 | 11.5 | Non-Mediterranean dry acid and neutral grassland |
| 5209 | 1 | 1 | 4 | 20 | 30 | 25 | Mesic grasslands without Mountain hay meadows |
| 5230 | 1 | 1 | 4 | 10 | 20 | 15 | Mountain hay meadows |
| 5300 | 1 | 1 | 11 | 10 | 25 | 17.5 | Seasonally wet and wet grasslands |
| 5400 | 1 | 2 | 5 | 5 | 10 | 7.5 | Alpine and subalpine grasslands |
| 6100 | 1 | 2 | 7 | 3 | 5 | 4 | Tundra |
| 6200 | 1 | 1 | 4 | 10 | 15 | 12.5 | Arctic alpine and subalpine scrub |
| 6400 | 1 | 3 | 17 | 10 | 20 | 15 | Temperate shrub heathland |
| 7100 | 4 | 7 | 15 | 10 | 20 | 15 | Broadleaved deciduous woodland |
| 7101 | 5 | 5 | 6 | 10 | 20 | 15 | Riparian Salix Alnus and Betula woodland |
| 7106 | 5 | 9 | 15 | 10 | 20 | 15 | Fagus woodland |
| 7201 | 4 | 7 | 10 | 3 | 7 | 5 | Mediterranean evergeeen [Quercus] woodland |
| 7300 | 3 | 5 | 9 | 10 | 20 | 15 | Coniferous woodland |
| 7301 | 3 | 6 | 10 | 10 | 15 | 12.5 | Abies and Picea woodland |
| 7302 | 5 | 6 | 9 | 10 | 20 | 15 | Alpine  Larix - Pinus cembra woodland |
| 7304 | 4 | 5 | 10 | 5 | 15 | 10 | Pinus sylvestris woodland south of the taiga |
| 7306 | 4 | 4 | 5 | 10 | 15 | 12.5 | Subalpine mediterranean Pinus woodland |
| 7400 | 3 | 5 | 11 | 5 | 20 | 12.5 | Mixed deciduous and coniferous woodland |
| 7401 | 4 | 6 | 11 | 5 | 20 | 12.5 | Mixed swamp woodland |
| 7402 | 4 | 4 | 9 | 5 | 8 | 6.5 | Mixed taiga woodland with Betula |
| 7403 | 4 | 6 | 10 | 5 | 20 | 12.5 | Mixed (sub)taiga woodland  acidophilous Quercus |
| 7404 | 4 | 4 | 9 | 5 | 20 | 12.5 | Mixed Pinus sylvestris - Betula woodland |
| 7406 | 4 | 4 | 8 | 10 | 20 | 15 | Mixed Abies - Picea - Fagus woodland |

| EUNIS | 5 | 50 | 95 | Empirical | | | Description |
|-------|---|----|----|-----------|-----|-----|-------------|
| | | | | min | max | avg | |
| 7407 | 4 | 6 | 9 | 5 | 20 | 12.5 | Mixed Pinus sylvestris – acid. Quercus woodland |
| 7411 | 6 | 6 | 6 | 5 | 20 | 12.5 | Mixed Mediterranean Pinus–therm. Quercus woodland |
| 7412 | 6 | 6 | 6 | 5 | 20 | 12.5 | Mixed Pinus sylvestris – therm. Quercus woodland |
| 7414 | 6 | 6 | 8 | 5 | 20 | 12.5 | Mixed Mediterranean pine - evergreen oak |

If the $N_{acc}$ criteria for vegetation change in western Europe are used, critical loads from SMB are much higher than when using the criteria for northern Europe; because leaching fluxes are much higher (Table 14).

**Table 14: SMB critical loads (5, 50 and 95 percentile) using the $[N]_{acc}$ criterium for western Europe and empirical critical loads (lower value of the range, upper value and average) in kg N.ha$^{-1}$**

| Eunis | 5 | 50 | 95 | Empirical | | | Description |
|-------|----|----|-----|-----------|-----|-----|-------------|
| | | | | min | max | avg | |
| 4100 | 10 | 25 | 97 | 5 | 10 | 7.5 | Raised and blanket bogs |
| 5120 | 1 | 5 | 12 | 15 | 25 | 20 | Perennial grasslands and basic steppes |
| 5179 | 2 | 8 | 146 | 8 | 15 | 11.5 | Non-Mediterranean dry acid and neutral grassland |
| 5209 | 3 | 11 | 46 | 20 | 30 | 25 | Mesic grasslands without Mountain hay meadows |
| 5230 | 3 | 15 | 46 | 10 | 20 | 15 | Mountain hay meadows |
| 5300 | 2 | 10 | 161 | 10 | 25 | 17.5 | Seasonally wet and wet grasslands |
| 5400 | 5 | 18 | 65 | 5 | 10 | 7.5 | Alpine and subalpine grasslands |
| 6100 | 10 | 29 | 97 | 3 | 5 | 4 | Tundra |
| 6200 | 4 | 14 | 48 | 10 | 15 | 12.5 | Arctic alpine and subalpine scrub |
| 6400 | 6 | 35 | 248 | 10 | 20 | 15 | Temperate shrub heathland |
| 7100 | 7 | 14 | 40 | 10 | 20 | 15 | Broadleaved deciduous woodland |
| 7101 | 7 | 8 | 19 | 10 | 20 | 15 | Riparian Salix Alnus and Betula woodland |
| 7106 | 10 | 19 | 39 | 10 | 20 | 15 | Fagus woodland |
| 7201 | 7 | 11 | 27 | 3 | 7 | 5 | Mediterranean evergeeen [Quercus] woodland |
| 7300 | 6 | 13 | 49 | 10 | 20 | 15 | Coniferous woodland |
| 7301 | 6 | 18 | 44 | 10 | 15 | 12.5 | Abies and Picea woodland |
| 7302 | 10 | 18 | 39 | 10 | 20 | 15 | Alpine Larix - Pinus cembra woodland |
| 7304 | 5 | 16 | 65 | 5 | 15 | 10 | Pinus sylvestris woodland south of the taiga |
| 7306 | 5 | 5 | 8 | 10 | 15 | 12.5 | Subalpine mediterranean Pinus woodland |

| Eunis | 5 | 50 | 95 | Empirical | | | Description |
|-------|---|----|----|-----------|---|---|-------------|
| | | | | min | max | avg | |
| 7400 | 8 | 15 | 51 | 5 | 20 | 12.5 | Mixed deciduous and coniferous woodland |
| 7401 | 11 | 24 | 91 | 5 | 20 | 12.5 | Mixed swamp woodland |
| 7402 | 6 | 10 | 30 | 5 | 8 | 6.5 | Mixed taiga woodland with Betula |
| 7403 | 6 | 11 | 32 | 5 | 20 | 12.5 | Mixed (sub)taiga woodland  acidiphilous Quercus |
| 7404 | 5 | 9 | 68 | 5 | 20 | 12.5 | Mixed Pinus sylvestris - Betula woodland |
| 7406 | 7 | 9 | 28 | 10 | 20 | 15 | Mixed Abies - Picea - Fagus woodland |
| 7407 | 6 | 10 | 20 | 5 | 20 | 12.5 | Mixed Pinus sylvestris – acid. Quercus woodland |
| 7411 | 7 | 8 | 8 | 5 | 20 | 12.5 | Mixed Mediterranean Pinus–therm. Quercus woodland |
| 7412 | 6 | 12 | 16 | 5 | 20 | 12.5 | Mixed Pinus sylvestris – therm. Quercus woodland |
| 7414 | 6 | 7 | 10 | 5 | 20 | 12.5 | Mixed Mediterranean pine - evergreen oak |

Median values for CLnutN compare quite well with the mean values of the empirical range (Figure 21), especially for forest ecosystems.

**Figure 21: Median values for CLnutN from EUDB 2020 (green) and the average empirical critical loads for N (blue) in kg.ha$^{-1}$.yr$^{-1}$ for the various EUNIS classes**
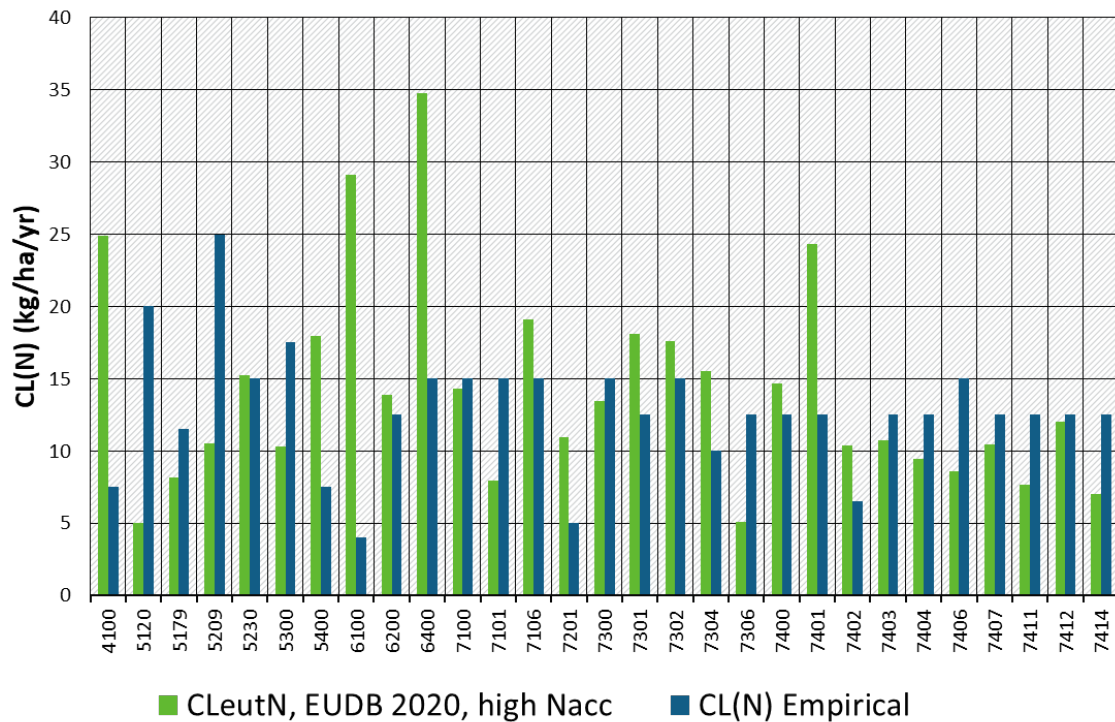


Source: Own representation, WUR,IIASA,RIVM

For some non-forest ecosystems, such as 'Alpine grasslands' and 'Tundras', the median CLnutN is much higher than the upper value of the empirical range. In high precipitation areas, the criterion for $N_{acc}$ of 2-3 mgN.l$^{-1}$ leads to unrealistically high critical loads, as can be seen from the 95 percentile values for, e.g., 'Seasonally wet and wet grasslands' and 'Temperate shrub heathland'.

# 9 Conclusions

A software package in R, EUDB 2020, was developed for UBA-CCE to compute critical loads of S and N for Europe. A comparison of results from EUDB 2020 with the original Fortran software used in the final RIVM-CCE report (Posch & Reinds, 2017) shows that results are almost identical. Some differences occur because of the update of hydrology and forest growth data in EUDB 2020.

The land use map used (Cinderby et al., 2007), meanwhile, is almost 15 years old. Due to land use changes that occurred in Europe over the past decades, an update of this land use map is advised.

The procedure now uses the most recent meteorological data. Although the R procedures allow for an update of these data, there is no need to do so on the short term unless the quality of the basic data would improve. Using updated data sets with more recent years will hardly change the results as the hydrology is computed and averaged over a twenty-year period.

UBA-CCE could consider making the R package BGDB with the basic critical load functions and transfer functions available to the National Focal Centres of the ICP M&M. With that, standardized and tested routines would be available for the use in national critical load computations.

# 10 References

Alexeyev, V.A., M.V. Markov & R.A. Birdsey, 2004. *Statistical Data on Forest Fund of Russia and Changing of Forest Productivity in the Second Half of XX Century*. St. Petersburg, Saint-Petersburg Forest Ecological Center.

Bobbink, R. & J.-P. Hettelingh, 2011. *Review and revision of empirical critical loads and dose-response relationships : Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010.* Bilthoven, Coordination Centre for Effects, National Institute for Public Health and the Environment. Report 680359002/2011.

Bonten, L.T.C., G.J. Reinds & M. Posch, 2016. *A model to calculate effects of atmospheric deposition on soil acidification, eutrophication and carbon sequestration.* Environ. Model. Softw. 79, 75-84.

Breeuwsma, A., J.P. Chardon, J.F. Kragt & W. de Vries, 1991. *Pedotransfer functions for denitrification.* In: Soil and Groundwater Research Report II: Nitrate in Soils. Luxembourg, Commission of the European Community, pp. 207-215.

Cinderby, S., L. Emberson, A. Owen & M. Ashmore, 2007. *LRTAP land cover map of Europe.* In: Slootweg J, P.M., Hettelingh J-P (Ed). Critical loads of nitrogen and dynamic modelling, CCE Progress Report 2007. . Bilthoven, the Netherlands, Report 500090001, MNP, pp. 59-70.

CLRTAP, 2017. *Mapping Manual. https://www.umweltbundesamt.de/sites/default/files/medien/4292/dokumente/ch5-mapman-2017-09-10.pdf.*

Davies, C.E. & D. Moss, 2002. *EUNIS Habitat Classification.* Centre for Ecology and Hydrology. 2001 Work Programme, Final Report to the European Environment Agency European Topic Centre on Nature Protection and Biodiversity.

De Vries, W., 1994. *Soil response to acid deposition at different regional scales; Field and laboratory data, critical loads and model predictions.* PhD Thesis, Wageningen, Wageningen University.

De Vries, W., M. Posch, H.U. Sverdrup, T. Larssen, H.A.d. Wit, R. Bobbink & J.-P. Hettelingh, 2015. *Geochemical indicators for use in the computation of critical loads and dynamic risk assessments.* Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems.

European Soil Bureau Network, 2004. *The European Soil Database distribution version 2.0.  CD ROM.*

FAO-UNESCO, 2003. *Digital soil map of the world and derived soil properties. CD-ROM, Rome, FAO.*

Gunn, J. & S.T. Trudgill, 1982. *Carbon dioxide production and concentrations in the soil atmosphere: a case study from New Zealand volcanic ash soils.* Catena 9, 81-94.

Harned, H.S. & R. Davis, 1943. *The ionisation constant of carbon dioxide in water and aqueous salt solutions from 0° to 50°.* Journal of the American Chemical Society 65, 2030-2037.

Hjellbrekke, A.-G. & L. Tarrason, 2001. *Mapping of Concentrations in Europe Combining Measurements and Acid Deposition Models.* Water Air Soil Pollut. 130 (1), 1529-1534.

Höhle, J. & N. Wellbrock, 2017. *Immobilisation of nitrogen in context of critical loads. Literature review and analysis of German, French and Swiss soil data. Umweltbundesamt Texte | 71/2017*

Jacobsen, C., P. Rademacher, H. Meesenburg & K.J. Meiwes, 2003. *Element contents in tree compartments - Literature study and data collection [in German].* Göttingen, Germany, Niedersächsische Forstliche Versuchsanstalt. Berichte des Forschungszentrums Waldökosysteme, Series B, Vol. 69.

Lee, D.S. & J.M. Pacyna, 1999. *An industrial emission inventory of calcium for Europe.* Atmos. Environ. 33, 1687-1697.

Monteith, J.L. & M. Unsworth, 1990. *Principles of Environmental Physics (2nd edition).* London, Arnold.

New, M., D. Lister, M. Hulme & I. Makin, 2002. *A high-resolution data set of surface climate over global land areas. .* Clim. Res. 21, 1-25.

Petz, K., C.J.E. Schulp, E.H.v.d. Zanden, C. Veerkamp, M.-J. Schelhaas, G.-J. Nabuurs & G. Hengeveld, 2016. *Indicators and modelling of land use, land management and ecosystem services.* PBL Netherlands Environmental Assessment Agency. Methodological documentation Nature Outlook.

Posch, M., W. de Vries & H.U. Sverdrup, 2015. *Mass Balance Models to Derive Critical Loads of Nitrogen and Acidity for Terrestrial and Aquatic Ecosystems.* In: De Vries, W., J.-P. Hettelingh & M. Posch (Eds). Critical Loads and Dynamic Risk Assessments. Vol. 25 Springer Netherlands, pp. 171-205.

Posch, M. & G.J. Reinds, 2017. *The European Background Database of N and S Critical Loads. .* In: Hettelingh, J.P., M. Posch & J. Slootweg (Eds). European critical loads: database, biodiversity and ecosystems at risk : CCE Final Report 2017. RIVM Report 2017-0155. Bilthoven, Netherlands, pp. 49-60.

Prentice, I.C., M.T. Sykes & W. Cramer, 1993. *A simulation model for the transient effects of climate change on forest landscapes.* Ecol. Model. 65 (1-2), 51-70.

Prins, A.G., R. Pouwels, J. Clement, H. Hendriks, B. de Knegt, K. Petz, A. Beusen, H. Farjon, A. van Hinsberg, J. Janse, O. Knol, P. van Puijenbroek, M. Schelhaas & S. van Tol, 2017. *Perspectives on the future of nature in Europe: impacts and combinations. PBL Netherlands Environmental Assessment Agency, The Hague. PBL publication number: 1784.*

Reinds, G.J., M. Posch & W. de Vries, 2001. *A semi-empirical dynamic soil acidification model for use in spatially explicit integrated assessment models for Europe.* Wageningen (Netherlands), Alterra. Alterra rapport 84.

Reinds, G.J., M. Posch, W. de Vries, J. Slootweg & J.P. Hettelingh, 2008a. *Critical loads of sulphur and nitrogen for terrestrial ecosystems in Europe and Northern Asia using different soil chemical criteria.* Water Air Soil Pollut. 193 (1), 269-287.

Reinds, G.J., M. van Oijen, G.B.M. Heuvelink & H. Kros, 2008b. *Bayesian calibration of the VSD soil acidification model using European forest monitoring data.* Geoderma 146 (3-4), 475-488.

Rosén, K., P. Gundersen, L. Tegnhammar, M. Johansson & T. Frogner, 1992. *Nitrogen enrichment of Nordic forest ecosystems - the concept of critical loads.* Ambio 21 (5), 364-368.

Santore, R.C., C.T. Driscoll & M. Aloi, 1995. *A model of soil organic matter and its function in temperate forest soil development.* In: McFee, W.W. & J.M. Kelly (Eds). Carbon forms and functions in forest soils. Madison, Wisconsin, Soil Sci. Soc. of America, pp. 275-298.

Schelhaas, M.J., S. Varis, A. Schuck & G.J. Nabuurs, 2006. *EFISCEN Inventory Database*. Joensuu, Finland, European Forest Institute. http://www.efi.int/databases/efiscen/

Schelhaas, M.J., J. Eggers, M. Lindner, G.J. Nabuurs, A. Pussinen, R. Päivinen, A. Schuck, P.J. Verkerk, D.C. van der Werf & S. Zudin, 2007. *Model documentation for the European Forest Information Scenario Model (EFISCEN 3.1.3).* Wageningen, Alterra. Alterra report 1559.

Schlutow, A., H.-D. Nagel & Y. Bouwer, 2017. *Bereitstellung der Critical Load Daten für den Call for Data 2015-2017 des Coordination Centre for Effects im Rahmen der Berichtspflichten Deutschlands für die Konvention über weitreichende grenzüberschreitende Luftverunreinigungen (CLRTAP).* Gesellschaft für Ökosystemanalyse und Umweltdatenmanagement mbH (ÖKO-DATA).

Slootweg, J., M. Posch & A. Warrink, 2009. *Status of the harmonised European land cover map.* In: Hettelingh J-P, P.M., Slootweg J (Ed). Progress in the modelling of critical thresholds, impacts to plant species diversity and ecosystem services in Europe : CCE Status Report 2009. Report 500090004. Bilthoven, the Netherlands, PBL, pp. 31-34.

Tegen, I. & I. Fung, 1995. *Contribution to the atmospheric mineral aerosol load from land surface modification.* J. Geophys. Res. 100, 18707-18726.

Van der Salm, C., 1999. *Weathering of forest soils.PhD dissertation. .* PhD dissertation, Amsterdam, University of Amsterdam.

Van Loon, M., L. Tarrasón & M. Posch, 2005. *Modelling base cations in Europe.* Oslo, Norway. EMEP Technical Report MSC-W 2/2005.

Vanmechelen, L., R. Groenemans & E. van Ranst, 1997. *Forest soil condition in Europe. Results of a Large-Scale Soil Survey*. Brussels, Geneva, EC-UN/ECE.

# A   Appendices

**A.1  Soil characteristics in the upper 50 cm of the soil as a function of soil type: median values for soil pH, soil organic carbon content, soil N content, soil carbonate content , and the number of observations the median value is based on.**

| Soiltype | parent material | Soil pH | Corg (%) | N (%) | CaCO3 (%) | #obs |
|---|---|---|---|---|---|---|
| A | 2 | 4.3 | 2.9 | 1.9 | 0 | 46 |
| Af | 2 | 4.6 | 4.5 | 2.5 | 0 | 13 |
| Ag | 2 | 3.8 | 2.5 | 1.6 | 0 | 7 |
| Ah | 1 | 4.4 | 7.2 | 4.6 | 0 | 4 |
| Ao | 1 | 4.3 | 2.3 | 1.5 | 0 | 22 |
| B | 1 | 4.2 | 2.6 | 1.8 | 0 | 1249 |
| Ba | 1 | 4.4 | 1.3 | 1.1 | 0 | 13 |
| Bc | 4 | 6 | 3.8 | 2.7 | 0 | 59 |
| Bd | 1 | 3.9 | 2.6 | 1.7 | 0 | 621 |
| Be | 1 | 4.5 | 2.4 | 1.7 | 0 | 246 |
| Bf | 1 | 3.9 | 2.9 | 1.5 | 0 | 8 |
| Bg | 4 | 3.8 | 2.1 | 1.3 | 0 | 66 |
| Bh | 1 | 4.1 | 4.6 | 3 | 0 | 66 |
| Bk | 4 | 7.3 | 3 | 2.3 | 120.8 | 147 |
| Bv | 2 | 4 | 1.8 | 0.9 | 0 | 11 |
| C | 2 | 5.9 | 1.9 | 2.1 | 0 | 11 |
| Ch | 2 | 5.9 | 0.7 | 1.6 | 0 | 7 |
| Ck | 4 | 6.8 | 1.9 | 2.1 | 26.3 | 3 |
| Cl | 2 | 5.5 | 3.2 | 3.1 | 0 | 1 |
| D | 1 | 3.9 | 1.9 | 1.1 | 0 | 48 |
| Dd | 1 | 3.9 | 1.5 | 0.7 | 0 | 10 |
| De | 1 | 3.8 | 1.2 | 0.7 | 0 | 3 |
| Dg | 1 | 3.8 | 2.2 | 1.4 | 0 | 35 |
| E | 4 | 7 | 4.5 | 3.1 | 24 | 286 |

| Soiltype | parent material | Soil pH | Corg (%) | N (%) | CaCO3 (%) | #obs |
|---|---|---|---|---|---|---|
| Ec | 4 | 7 | 6.4 | 4.4 | 40 | 130 |
| G | 2 | 4.2 | 3 | 1.8 | 0 | 135 |
| Gc | 4 | 6.6 | 4.5 | 3.1 | 1 | 10 |
| Gd | 2 | 3.8 | 2 | 1.2 | 0 | 54 |
| Ge | 2 | 4.3 | 2.8 | 1.9 | 0 | 46 |
| Gh | 2 | 3.9 | 4.1 | 2 | 0 | 21 |
| Gm | 2 | 4.8 | 4.4 | 3.4 | 0 | 4 |
| H | 2 | 6.6 | 3.8 | 3.1 | 16 | 9 |
| Hc | 4 | 6.6 | 3.8 | 3.2 | 16 | 6 |
| Hh | 2 | 6 | 1.3 | 1.3 | 4.3 | 1 |
| Hl | 2 | 6.6 | 1.6 | 1.8 | 1.5 | 2 |
| I | 1 | 4.7 | 3.8 | 2.1 | 0 | 63 |
| J | 2 | 7 | 2 | 1.5 | 0 | 29 |
| Jc | 4 | 7.5 | 1.4 | 1.1 | 42.3 | 15 |
| Jd | 1 | 3.9 | 1.9 | 1.5 | 0 | 4 |
| Je | 2 | 5.8 | 2.3 | 1.8 | 0 | 10 |
| K | 4 | 7.6 | 3.6 | 3 | 493.5 | 10 |
| Kh | 4 | 7.5 | 4.7 | 3.1 | 426.5 | 8 |
| Kk | 4 | 7.6 | 2.9 | 1.7 | 493.5 | 2 |
| L | 2 | 4.2 | 2 | 1.5 | 0 | 408 |
| La | 2 | 3.7 | 2.4 | 1.9 | 0 | 38 |
| Lc | 4 | 5.7 | 2.5 | 1.8 | 0 | 54 |
| Ld | 2 | 4.5 | 1.8 | 1.1 | 0 | 17 |
| Lg | 2 | 4.2 | 2.1 | 1.5 | 0 | 128 |
| Lk | 4 | 4.2 | 2.2 | 1.6 | 0 | 6 |
| Lo | 2 | 4.2 | 1.7 | 1.4 | 0 | 162 |
| Lv | 4 | 5.2 | 3.8 | 2.8 | 0 | 3 |
| Mo | 2 | 3.9 | 1.1 | 0.6 | 0 | 4 |
| O | 0 | 3.5 | 41.4 | 14.3 | 0 | 191 |

| Soiltype | parent material | Soil pH | Corg (%) | N (%) | CaCO3 (%) | #obs |
|----------|-----------------|---------|----------|-------|-----------|------|
| P | 1 | 4 | 2.3 | 0.9 | 0 | 1189 |
| Pf | 1 | 4.1 | 1.4 | 0.6 | 0 | 270 |
| Pg | 1 | 3.8 | 2.5 | 1 | 0 | 83 |
| Ph | 1 | 3.9 | 3.3 | 1.2 | 0 | 138 |
| Pl | 1 | 3.9 | 3.5 | 1.7 | 0 | 153 |
| Po | 1 | 3.9 | 2.3 | 0.9 | 0 | 545 |
| Q | 1 | 4 | 1.6 | 0.7 | 0 | 586 |
| Qa | 1 | 3.8 | 1.1 | 0.5 | 0 | 9 |
| Qc | 1 | 4 | 1.4 | 0.8 | 0 | 192 |
| Qh | 1 | 3.9 | 1.9 | 0.7 | 0 | 365 |
| Ql | 1 | 3.9 | 0.6 | 0.6 | 0 | 16 |
| R | 2 | 4.3 | 2.1 | 1.2 | 0 | 261 |
| Rc | 4 | 7.5 | 1.9 | 1.7 | 336.8 | 44 |
| Rd | 1 | 4.1 | 2.2 | 1 | 0 | 187 |
| Re | 2 | 4.5 | 2.2 | 1.1 | 0 | 30 |
| S | 4 | 7.9 | 2.5 | 4.7 | 682.5 | 1 |
| Sm | 4 | 7.9 | 2.5 | 4.7 | 682.5 | 1 |
| T | 3 | 4.7 | 4.1 | 3.7 | 0 | 10 |
| Th | 3 | 5.1 | 6.3 | 3.7 | 0 | 1 |
| To | 3 | 4.6 | 4.7 | 4.6 | 0 | 5 |
| Tv | 3 | 6.4 | 0.7 | 1.2 | 0 | 4 |
| U | 1 | 4.2 | 3.7 | 2.1 | 0 | 173 |
| Ud | 1 | 4.2 | 2.7 | 1.8 | 0 | 113 |
| Uk | 4 | 4.2 | 6.8 | 3.6 | 0 | 37 |
| V | 2 | 4.7 | 1.1 | 1.5 | 0 | 10 |
| Vc | 4 | 5.2 | 2 | 1.8 | 0 | 7 |
| Vp | 2 | 3.8 | 1 | 0.6 | 0 | 3 |
| W | 2 | 3.7 | 1.9 | 1.1 | 0 | 63 |
| Wd | 1 | 3.9 | 1.5 | 1 | 0 | 41 |

| Soiltype | parent material | Soil pH | Corg (%) | N (%) | CaCO3 (%) | #obs |
|----------|-----------------|---------|----------|-------|-----------|------|
| We | 2 | 3.7 | 2.6 | 1.2 | 0 | 15 |

## A.2  Documentation of the package BGDB

See separate PDF document

## A.3  Conversion of radiation data

The downloaded daily radiation data in the EObs data set are called 'surface downwelling shortwave flux in air' and given in W/m². In Figure 22 (left) these are shown for the 0.5°×0.5° grid cell holding Wageningen (SW-corner: 5.5°E, 51.5°N) for the years 1999, 2009 and 2018. Assuming that it represents the daily (=24 hour) average, one has to multiply by 24×3600 to get the total daily radiation in J/m²/day (remember: W = J/s). Doing so, one obtains the values shown in Figure 22 (right; in MJ/m²/day).

**Figure 22: Daily solar downward radiation in W/m² at the Wageningen grid cell: (a) for the years 1999 (blue), 2009 (green), and 2018 (red); (b) the daily 20-year (1999-2018) minimum and maximum daily downward radiation, expressed in MJ/m²/. The red and orange curves show the daily radiation for $n_j$=0 (no sunshine) and $n_j$=1 (=100% sunshine) as computed by eq.B4 (see below)**
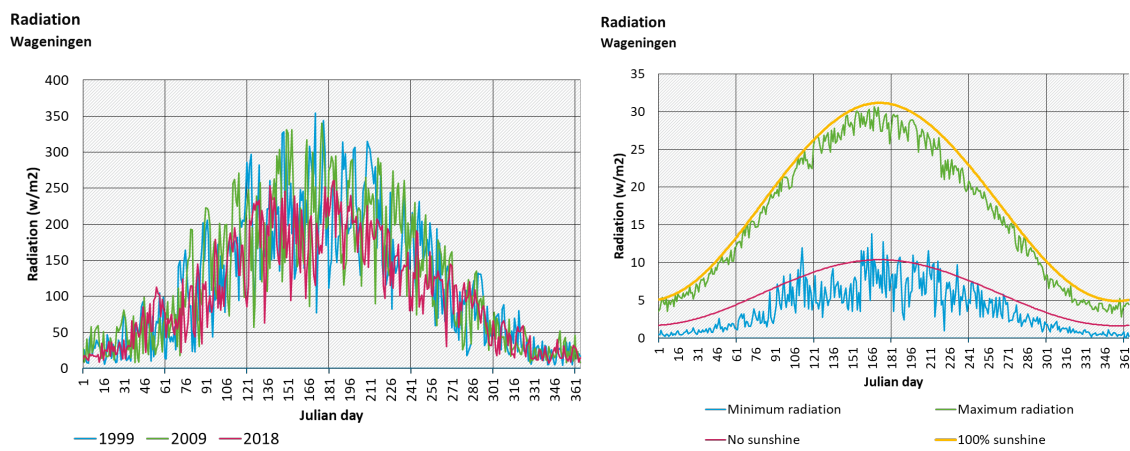


In MetHyd the 'instantaneous' net downward shortwave flux $R_{s,t}$ (in W/m²) for zero albedo (i.e. nothing reflected) is given as (see eq.P3 in MHM[=MetHyd Manual], Bonten et al. (2016)):

(B1)    $R_{s,t} = (c + dn_j)S_j \cos z_j$

Where $n_j$ is the fraction of sunshine on day $j$ ($j$=1,365), $c$=0.25 and $d$=0.5 are (empirical) constants ($c$+$d$ is the clear-sky transmissivity), $S_j$ is the solar constant (1360.8 W/m², if taken independent of the day) and $z_j$ is zenith angle of the Sun (i.e. the angle measured from the vertical); this is given as (see eq.R2 in MHM):

(B2)    $\cos z_j = \sin \lambda \sin \delta_j + \cos \lambda \cos \delta_j \cos(\omega t)$

Where $\lambda$ is the longitude of the site, $\delta_j$ is the solar declination on day $j$, and $t$ the time of the day measured from noon, and $\omega$=2π/24 if $t$ is measured in hours [$\omega$=2π/(24·3600) if $t$ is measured in seconds, etc.]. The time $t_j$ from solar noon to sunset is found by setting $z_j$=π/2 (i.e. cos$z_j$=0) in eq.B2 and solving it to obtain:

(B3)    $t_j = \frac{1}{\omega} \arccos(-\tan \lambda \tan \delta_j)$

75

If the argument of the *arccos* in eq.B3 is > 1, then there is polar night ($t_j$=0), if it is < –1 there is midnight sun ($t_j$=12). The total daily radiation at day $j$, $R_{s,j}$, is obtained by integrating $R_{s,t}$ over the whole day, i.e. between $-t_j$ and $t_j$, obtaining:

(B4)  $R_{s,j} = (c + dn_j)S_jI_j$

where

(B5a)  $I_j = \int_{-t_j}^{t_j} \cos z_j \, dt = 2\left(s_jt_j + \frac{c_j}{\omega}\sin \omega t_j\right)$

with

(B5b)  $s_j = \sin \lambda \sin \delta_j$  and  $c_j = \cos \lambda \cos \delta_j$

The quantity Rs,j was compared for nj=1 (no clouds at all) at the Wageningen cell. Results showed that it fits reasonably well with the maxima of the observations. One has to bear in mind that the parameters *c* and *d* in eq.B1 are 'empirical' (from Prentice et al. (1993)), i.e. changing them could lead to a better fit.

For computing evapotranspiration we need the instantaneous radiation (eq.B1). To make use of the measured radiation we thus set:

(B6)  $R_{s,t} = \left(1 - a_j\right)\frac{24R_{s,meas}}{I_j}\cos z_j$

Where $R_{s,meas}$ is the measured daily radiation (in W/m²) and $I_j$ given by eq.B5. We also multiplied with $1-a_j$ ($a_j$ = short-wave albedo), to get the net radiation to the ground. The factor 24 (h/day) arises from the fact that $I_j$ has dimension 'hours'. Integrating eq.B6 over the daylength [$-t_j,t_j$] we obtain, for zero albedo. $R_{s,t} = R_{s,meas}$.

For computing evapotranspiration we also need the 'instantaneous' net upward long-wave flux $R_{l,t}$ (W/m²). In MetHyd this is approximated by a linear function of the temperature $T$ (in °C) (see eq.P4 in MHM; see also Monteith and Unsworth (1990)):

(B7)  $R_{l,t} = [b + (1 - b)n_j](A - aT)$

where $b$=0.2, $A$=107 W/m², and $a$=1 W/m²/°C are empirical constants. Taking $T$ as the mean daily temperature we neglect the effects of diurnal temperature variations on $R_{l,t}$.

The problem is that in eq.B7 the sunshine fraction is needed! A possible solution is to set $R_{s,j}$ in eq.B4 to $R_{s,meas}$ and solve for $n_j$, i.e.:

(B8)  $n_j = \min\left\{\max\left\{0, \left(\frac{24R_{s,meas}}{s_jI_j} - c\right)/d\right\}, 1\right\}$

where the 'min-max' takes care that 0≤$n_j$≤1. Care has also to be taken in case $I_j$=0 (polar nights), but then 'sunshine' is not of relevance.

## A.4  Area (in km$^2$) and number of receptors per country and EUNIS class in the critical load database

| Country | EUNIS E | | EUNIS D | | EUNIS F | | EUNIS G | |
|---|---|---|---|---|---|---|---|---|
| | area | Number of receptors | area | Number of receptors | area | Number of receptors | area | Number of receptors |
| AL | 7678 | 26784 | 24 | 104 | 4869 | 16344 | 7447 | 21588 |
| AT | 15023 | 34647 | 209 | 577 | 2520 | 6142 | 37198 | 72650 |
| BA | 11071 | 35406 | 47 | 128 | 3318 | 11051 | 22668 | 63000 |
| BE | 6437 | 18558 | 85 | 289 | 151 | 530 | 6175 | 19140 |
| BG | 15191 | 57410 | 99 | 383 | 1863 | 5602 | 37317 | 94568 |
| BY | 3941 | 6474 | 2825 | 3257 | 124 | 250 | 59723 | 70463 |
| CH | 7884 | 15951 | 0 | 0 | 1879 | 4086 | 13225 | 25838 |
| CZ | 8703 | 32939 | 87 | 422 | 26 | 128 | 26767 | 83605 |
| DE | 55566 | 169735 | 1320 | 5147 | 625 | 2285 | 103910 | 303124 |
| DK | 1869 | 8909 | 564 | 2858 | 471 | 1426 | 3836 | 17017 |
| EE | 7516 | 22854 | 1354 | 3102 | 120 | 356 | 21745 | 55389 |
| ES | 100996 | 262703 | 562 | 811 | 59188 | 145945 | 91704 | 219831 |
| FI | 44645 | 180499 | 22152 | 91646 | 9573 | 20307 | 189707 | 558914 |
| FR | 167257 | 306064 | 747 | 1527 | 9968 | 20623 | 145306 | 300577 |
| GB | 95145 | 195367 | 5201 | 12594 | 27772 | 57962 | 19203 | 61408 |
| GR | 32295 | 121634 | 196 | 860 | 20972 | 66232 | 24552 | 82521 |
| HR | 14256 | 40811 | 161 | 441 | 2261 | 7338 | 19806 | 46272 |
| HU | 11715 | 36003 | 997 | 3174 | 0 | 0 | 17407 | 44120 |
| IE | 43815 | 67429 | 11351 | 21412 | 573 | 1186 | 2917 | 9079 |
| IT | 35275 | 116677 | 141 | 480 | 5906 | 22511 | 78921 | 199518 |
| LI | 35 | 71 | 0 | 0 | 11 | 30 | 63 | 137 |
| LT | 7082 | 33470 | 546 | 2205 | 40 | 148 | 18855 | 74975 |
| LU | 460 | 1473 | 0 | 0 | 0 | 0 | 928 | 2871 |
| LV | 15708 | 63306 | 1454 | 3721 | 0 | 0 | 27226 | 102122 |
| MD | 1838 | 1514 | 0 | 0 | 111 | 291 | 1831 | 1962 |

| | EUNIS E | | EUNIS D | | EUNIS F | | EUNIS G | |
|---|---|---|---|---|---|---|---|---|
| MK | 6375 | 25622 | 12 | 68 | 2193 | 8369 | 8342 | 24903 |
| NL | 12664 | 21481 | 355 | 666 | 348 | 1323 | 2869 | 10022 |
| NO | 8716 | 23304 | 701 | 1532 | 172197 | 275472 | 84501 | 206105 |
| PL | 33395 | 148587 | 1065 | 4997 | 137 | 593 | 93071 | 362475 |
| PT | 14508 | 69035 | 9 | 46 | 4684 | 20640 | 22998 | 94872 |
| RO | 34216 | 83515 | 9 | 20 | 3640 | 10663 | 71698 | 129380 |
| RS | 12876 | 36073 | 0 | 0 | 410 | 1739 | 19835 | 57474 |
| RU | 110159 | 110495 | 0 | 0 | 14425 | 14861 | 519302 | 489240 |
| SE | 42996 | 244711 | 27264 | 159598 | 26675 | 87159 | 247366 | 773002 |
| SI | 2778 | 7333 | 10 | 50 | 307 | 859 | 11009 | 23446 |
| SK | 4591 | 19748 | 41 | 188 | 1532 | 7649 | 20712 | 67401 |
| UA | 22194 | 19900 | 0 | 0 | 1151 | 824 | 74569 | 82138 |
| total | 1016869 | 2666492 | 79588 | 322303 | 380040 | 820924 | 2154709 | 4851147 |