> Critical Loads of Nitrogen and their Exceedances

Swiss contribution to the effects-oriented work under the Convention on Long-range Transboundary Air Pollution (UNECE)

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> Abstracts

Critical loads are defined as those air pollutant depositions below which harmful effects on specified sensitive receptors of the environment do not occur according to present scientific knowledge. They are an established element for developing effects-based approaches under the UNECE Convention on Long-range Transboundary Air Pollution. Critical loads of nitrogen were determined and mapped for forests and for (semi-)natural ecosystems in Switzerland by applying two methods proposed by the UNECE: the simple mass balance (SMB) and the empirical method. Total nitrogen deposition was modelled with high spatial resolution for the time period 1990–2010. In 2010, the nitrogen deposition exceeded the critical loads on more than 90% of all forest sites and on approximately 70% of the (semi-)natural ecosystems.

Keywords:
Critical loads of nitrogen, (semi-) natural ecosystems, nitrogen deposition, exceedance of critical loads, UNECE LRTAP Convention

Als Critical Loads werden jene Depositionen oder Einträge von Luftschadstoffen bezeichnet, unterhalb welchen nach dem heutigen Stand des Wissens keine schädlichen Auswirkungen auf empfindliche Rezeptoren der Umwelt auftreten. Im Rahmen der Konvention über weiträumige grenzüberschreitende Luftverunreinigung (UNECE) sind die Critical Loads wichtige Zielgrössen zur Entwicklung von wirkungsorientierten Luftreinhaltestrategien. Critical Loads für Stickstoff wurden in der Schweiz für Wälder und (halb)natürliche Ökosysteme mit zwei von der UNECE vorgeschlagenen Methoden bestimmt und kartiert: die Massenbilanzmethode (SMB) und die empirische Methode. Die Deposition von stickstoffhaltigen Luftschadstoffen wurde für den Zeitraum 1990–2010 in einer hohen räumlichen Auflösung modelliert. Im Jahr 2010 wurden die Critical Loads für Stickstoff bei mehr als 90% der Waldökosysteme und bei rund 70% der (halb)natürlichen Ökosysteme überschritten.

Stichwörter:
Critical Loads für Stickstoff, (halb)natürliche Ökosysteme, Stickstoffdeposition, Überschreitung der Critical Loads, UNECE LRTAP Convention

La notion des charges critiques représente une estimation quantitative des dépôts de polluants atmosphériques au-dessous desquels, selon les connaissances actuelles, il n'y a pas d'effets nocifs pour des milieux sensibles de l'environnement. L'approche basée sur les charges critiques est un élément essentiel de la Convention sur la pollution transfrontière à longue distance (CEE ONU) pour le développement de stratégies de lutte contre la pollution de l'air basées sur les effets. En Suisse, les charges critiques pour l'azote ont été déterminées et cartographiées pour les forêts et les écosystèmes (semi-)naturels en appliquant deux méthodes recommandées par la CEE ONU: le «bilan à l'équilibre» (SMB) et une méthode dite empirique. Les dépôts de composés azotés atmosphériques ont été modélisés pour la période 1990–2010 avec une résolution spatiale élevée. En 2010, ces dépôts dépassaient les charges critiques pour l'azote sur plus que 90% des sites forestiers et dans environ 70% des écosystèmes (semi-) naturels.

Mots-clés:
Charges critiques pour l'azote, écosystèmes (semi-)naturels, dépôts de composés azotés, dépassement de charges critiques, UNECE LRTAP Convention

La nozione dei carichi critici rappresenta una stima quantitativa dei depositi di inquinanti atmosferici al di sotto dei quali, secondo le conoscenze attuali, non ci sono effetti nocivi per i ricettori ambientali particolarmente sensibili. L'approccio basato sui carichi critici è un elemento essenziale della Convenzione sull'inquinamento atmosferico transfrontaliero a grande distanza (CEE ONU) per lo sviluppo di strategie di lotta contro l'inquinamento dell'aria basata sugli effetti. In Svizzera, i carichi critici per l'azoto sono stati determinati e mappati per le foreste e gli ecosistemi (semi-)naturali applicando due metodi raccomandati dalla CEE ONU: il metodo del "bilancio a l'equilibrio" (Simple Mass Bilance) e il metodo detto empirico. I depositi di composti azotati atmosferici sono stati modellizzati per il periodo 1990–2010 con una risoluzione spaziale elevata. Nel 2010, questi depositi oltrepassavano i carichi critici per l'azoto su più di 90% dei siti forestali e nel circa 70% degli ecosistemi (semi-)naturali.

Parole chiave:
Carichi critici per l'azoto, ecosistemi (semi-)naturali, depositi di composti azotati, superamento dei carichi critici, UNECE LRTAP Convention

> Foreword

A critical load is defined as “a quantitative estimate of an exposure to one or more air pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge”. The critical loads approach was developed under the Convention on Long-range Transboundary Air Pollution (UNECE LRTAP Convention) and its Working Group on Effects to support effects-oriented air pollution abatement policies. High atmospheric deposition of sulphur and nitrogen compounds and their harmful effects on sensitive ecosystems in terms of acidification and eutrophication were at the origin of substantial scientific efforts to derive ecosystem-specific critical loads of acidity and critical loads of nutrient nitrogen. To enable the long-term protection of ecosystems, the critical loads should not be exceeded by atmospheric deposition of sulphur and nitrogen. Many scientific workshops were held under the Convention since 1988 aiming at setting critical loads reflecting the most recent scientific knowledge on effects of air pollutants. The UNECE Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends summarizes these scientific findings and gives guidance to Parties to the Convention on how to apply the critical loads approach. Today, the critical loads and levels approach is an important element in the structure of the Gothenburg Protocol negotiated under the LRTAP Convention to abate acidification, eutrophication and ground-level ozone. According to the objective of this Protocol, the emissions of sulphur, nitrogen oxides, ammonia, volatile organic compounds and particulate matter shall be reduced to ensure that, in the long-term and in a step-wise approach, atmospheric depositions or concentrations do not exceed critical loads of acidity, critical loads of nutrient nitrogen, critical levels of ozone, critical levels of particulate matter, critical levels of ammonia and acceptable levels of air pollutants to protect materials.

Swiss scientists and experts have actively participated in the further development of the critical loads and levels approach under the LRTAP Convention. The report presented here focuses on the critical loads of nutrient nitrogen and their exceedances by elevated atmospheric deposition of nitrogen compounds on sensitive (semi-)natural ecosystems. It mainly reflects the scientific knowledge resulting from the workshops on critical loads of nitrogen held under the auspices of the Convention in Lökeberg (1992), in Berne (2002) and in Noordwijkerhout (2010), and it considers as well results from experimental work in natural ecosystems and from gradient studies carried out by scientists in Switzerland. The report is an update of the report on critical loads of nitrogen published by the Federal Office in 1996 (Environmental Series No. 275).

We are convinced that the ongoing work on critical loads and levels has the potential to contribute to increasing the quality and ambition level of future international agreements on the control of air pollutant emissions and also to the further development of national and regional air pollution control policies on the basis of effects-oriented assessments of impacts of air pollutants on sensitive ecosystems.

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> Summary

The negotiations on air pollution abatement strategies under the UNECE Convention on Long-range Transboundary Air Pollution are based on an effects-based critical levels / loads approach. Critical levels / loads are defined as those air pollutant concentrations / depositions below which harmful effects on specified sensitive receptors of the environment do not occur according to present scientific knowledge.

Under the Convention, the International Cooperative Programme (ICP) on "Modelling and Mapping Critical Levels and Loads and Air Pollution Effects, Risks and Trends" aims at developing scientific methods to compute and map the ecosystem sensitivities to air pollutants in the ECE region. A regularly updated Manual on Methodologies and Criteria for Mapping Critical Loads and Levels gives guidance to the Parties to the Convention to apply the most appropriate methods to assess the exposure of the ecosystems on their territory and the associated risks. At present, more than 20 countries are taking part in the ICP. Most of the countries supply national data and maps to the Coordination Centre for Effects (CCE), where the European maps are compiled and overall risk assessments are carried out.

For Switzerland, critical loads of nitrogen were computed using two methods proposed by the ICP: (1) the simple (steady state) mass balance method (SMB) for forests; and (2) the empirical method for natural and semi-natural ecosystems such as raised bogs, fens, dry or species-rich grassland, mountain hay meadows, (sub)alpine grassland, alpine scrub habitats and alpine lakes.

The SMB balances the atmospheric depositions against the natural long-term processes that permanently immobilise nitrogen or remove it from the (eco-)system. The SMB was applied on a 1 x 1 km² grid, with 10 632 points representing productive forests, i.e. sites where harvesting is possible.

The empirical method is based on results from scientific field studies with nitrogen addition experiments, from field observation studies along nitrogen deposition gradients, from mesocosm studies, but also on expert judgement. The result is a list of sensitive ecosystem types in Europe with corresponding ranges of approved critical load values. Swiss critical load maps were produced by assigning appropriate values from that list to the national inventories of ecosystems worthy to be protected (raised bogs, fens, dry grasslands (TWW), various ecosystems listed in the Swiss Atlas of Vegetation Types Worthy of Protection, data set of the Swiss Biodiversity Monitoring). For the use of the maps under the Convention, the maps were aggregated to a 1 x 1 km² raster with a total area of 18 584 km² containing sensitive ecosystems.

Nitrogen deposition in 1990, 2000 and 2010 was mapped using a pragmatic approach that combines emission inventories, statistical dispersion models, monitoring data, spatial interpolation methods and inferential deposition models. The following compounds were considered: wet and dry deposition of nitrate (NO_3^-) and ammonium

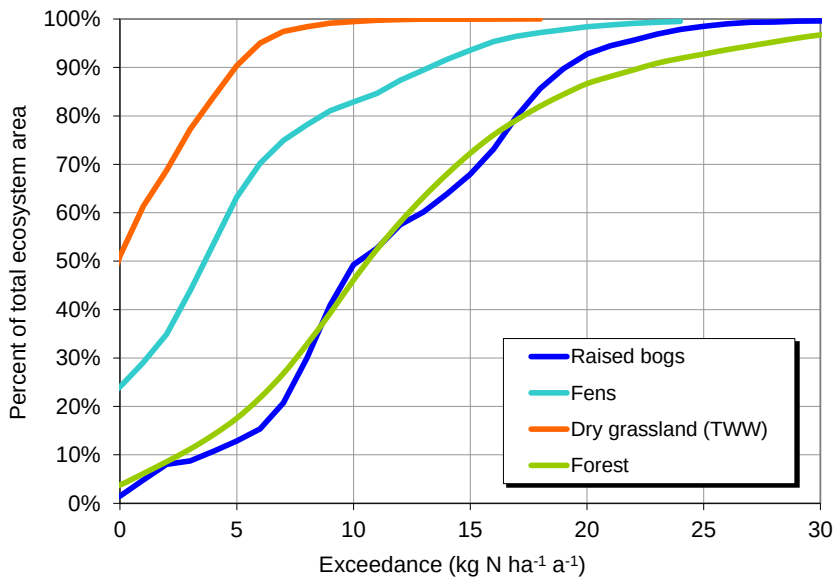
(NH_4^+) as well as gaseous deposition of ammonia (NH_3), nitrogen dioxide (NO_2) and nitric acid (HNO_3).

The resulting critical loads of nitrogen show a variation of 4 to 25 kg N ha^{-1} a^{-1}, depending on ecosystem type, soil type, altitude and other ecosystem properties. Nitrogen depositions in 2010 were in the range of 2 to 65 kg N ha^{-1} a^{-1}, the highest values occurring in the lowlands. The total deposition in Switzerland amounts to 67 kt N a^{-1} (44 kt N a^{-1} of reduced nitrogen plus 23 kt N a^{-1} of oxidised nitrogen). From 1990 to 2010 the total deposition of nitrogen compounds decreased by 23%.

At 95% of the forest sites the critical loads of nitrogen are exceeded in 2010 (Figure 1). The critical loads for (semi-)natural ecosystems derived by the empirical method and aggregated to the 1 x 1 km² raster are exceeded on 69% of the total sensitive area. A more detailed analysis on a 100 x 100 m² raster was made for the Swiss nature reserves; Figure 1 shows the resulting exceedances for the protected areas of raised bogs (98% of area exceeded), fens (76% of area exceeded) and species rich dry grassland TWW (49% of area exceeded).

Fig. 1 > Cumulative frequency distribution of the exceedances of critical loads of nutrient nitrogen for different protected ecosystems

Units: percent of total ecosystem area. Nitrogen deposition: year 2010.



In the case of forests, exceedances indicate a long-term risk of nutrient imbalances, growth disturbances (e.g. root / shoot ratio), changes in ground flora and mycorrhizas, increased nitrogen leaching and loss of stability of the forest. In the case of natural and semi-natural ecosystems, exceedances indicate a risk of changes in species composition and subsequent loss of biodiversity. The situation is less severe for (sub-) alpine grassland, alpine scrub habitats and forests in the inner-alpine valleys (Valais, Grisons), where exceedances are relatively small.

An important step to improve this situation will be made by achieving the 2020 emission reduction targets of the Gothenburg Protocol as revised in 2012. Further emission reductions under this international agreement beyond 2020 are possible and would result in substantial benefits for the environment.

1 > Background and Aims

1.1 The Critical Loads and Levels approach developed under the UNECE Convention on Long-range Transboundary Air Pollution

Since 1979 the Convention on Long-range Transboundary Air Pollution (LRTAP Convention, UNECE[1]) has addressed several environmental problems of the ECE region through scientific collaboration and policy negotiation. A good scientific understanding of the harmful effects of air pollution was recognised as a prerequisite for reaching political agreement on targeted pollution control. To develop the necessary international cooperation in the research on and the monitoring of pollutant effects, the Working Group on Effects (WGE) along with Task Forces and International Cooperative Programmes (ICPs) were established under the Convention. The EMEP[2] programme provides support to the Convention in the field of atmospheric transport modelling of pollutants.

At the sixth session of the Executive Body for the Convention in 1988, a Task Force on Mapping Critical Levels / Loads was established, with the aim of developing the mapping approaches needed to show the extent of the sensitivity of ecosystems to air pollution and the exceedances of critical levels / loads in the ECE region. Later the Task Force was renamed to 'ICP Modelling and Mapping of Critical Loads & Levels and Air Pollution Effects, Risks and Trends' (ICP Modelling & Mapping or ICP M&M[3]).

Definition of the critical load

The basic idea of the critical load concept is to balance the depositions which an ecosystem is exposed to with the capacity of this ecosystem to buffer the input (e.g. the acidity input buffered by the weathering rate), to remove it from the system (e.g. nitrogen by plant uptake and harvest) or to immobilize it in the long-term without harmful effects within or outside the system. Accordingly, a critical load is defined as "a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" (Nilsson & Grennfelt 1988, UNECE 2012). In the case of eutrophying effects of nitrogen the term "exposure" relates to the sum of the depositions of reduced nitrogen compounds (NH_x) and oxidised nitrogen compounds (NO_y). "Sensitive elements of the environment" can be whole ecosystems or part of them by addressing ecosystem development processes, structure and function (UNECE 2016, Chapter V.1).

The work programme under the ICP Modelling & Mapping includes the production of maps of critical loads, critical levels and their exceedances as a basis for developing effects-based abatement strategies for transboundary air pollutants. Several scientific

[1] www.unece.org/env/lrtap/welcome.html
[2] www.emep.int
[3] www.icpmapping.org

workshops were held under the auspices of the UNECE to define critical levels and critical loads for sensitive receptors of the environment and to discuss the methods for mapping them. Critical levels have been defined for the direct effects of ambient concentrations of SO_2, NO_x (NO and NO_2), NH_3 and O_3 on different vegetation types and on materials. Methods for the computation of critical loads of acidity (sulphur and nitrogen), of nutrient nitrogen with respect to eutrophication and of heavy metals have been proposed for aquatic and terrestrial ecosystems.

Scientific workshops addressing critical loads of nitrogen

With respect to critical loads of nutrient (eutrophying) nitrogen the following scientific workshops and meetings were held under the Convention:

- Workshop on critical loads for sulphur and nitrogen in Skokloster, Sweden, 1988 (Nilsson & Grennfelt 1988);
- Workshop on critical loads for nitrogen in Lökeberg, Sweden, 1992 (Grennfelt & Thörnelöf 1992);
- WHO workshop on Air Quality Guidelines for Europe, held in Les Diablerets, Switzerland (WHO/Europe 1995);
- Workshop on critical loads for nitrogen in Grange-over-Sands, United Kingdom, 1994 (Hornung et al. 1995);
- Meeting of the Task Force on Mapping with expert pre-meetings on empirical critical loads of nitrogen in Geneva, Switzerland, December 1995 (UNECE 1995);
- Expert Workshop on empirical critical loads for nitrogen, Berne, Switzerland, 11–13 November 2002 (Achermann & Bobbink 2003);
- Expert Workshop on review and revision of empirical critical loads and dose-response relationships, Noordwijkerhout, The Netherlands, 23–25 June 2010 (Bobbink & Hettelingh 2011).

Several workshops and meetings of the ICP Modelling & Mapping have led to the production and repeated revision of a “Manual on Methodologies for Mapping Critical Levels / Loads and Geographical Areas where they are Exceeded” (UNECE 1993, 1996, 2004 and 2016). The manual supplies a scientific basis and guidelines for gathering, handling and processing data on the basis of which Parties to the Convention are able to:

- determine and identify sensitive receptors and locations;
- map critical levels and loads on a national scale;
- map areas where air pollutant concentrations or depositions exceed critical levels or loads.

Coordination Centre for Effects

In order to provide scientific and technical assistance to the Task Force on Modelling & Mapping and to National Focal Centres (NFCs), a Coordination Centre for Effects (CCE[4]) was established at the National Institute for Public Health and the Environment (RIVM) in Bilthoven, The Netherlands. Since 1990 the CCE organizes annual training sessions for national mapping experts and for dynamic modellers, where technical aspects of the modelling and mapping approaches are discussed and coordinated.

[4] http://wge-cce.org

The CCE produces the “best available” integrated European critical levels / loads and exceedance maps, based primarily upon data supplied by individual countries and by EMEP. Since 1991, it also publishes technical reports on modelling and mapping covering topics such as critical loads of sulphur and nitrogen and their exceedances, dynamic modelling of soil processes, effects on ecosystems and biodiversity (e.g. Hettelingh et al. 1991, Posch et al. 2012, Slootweg et al. 2015).

National Focal Centres

In order to ensure the coordination of the national mapping activities, the Parties to the Convention were invited to establish National Focal Centres (NFCs). The Swiss National Focal Centre is located at the Federal Office for the Environment (FOEN).

With regard to critical loads the most important activities under the Convention on LRTAP can be summarised as follows.

Critical loads of acidity

According to the work plan of the Executive Body for the Convention, the first maps to be produced at the European scale in 1991/1992 were the map on critical loads of acidity and the map on critical loads of sulphur derived from it. Both maps were successfully used as a basis for developing effects-based and cost-optimized sulphur emission abatement scenarios by the UNECE Task Force on Integrated Assessment Modelling[5]. The scenario results have been used by the UNECE Working Group on Strategies in the negotiation process for the Protocol on the further reduction of sulphur emissions, which was signed in Oslo, Norway, in June 1994.

Critical loads of nitrogen

A second step under the Convention was the revision of the 1988 Protocol concerning the Control of Nitrogen Oxides. Critical loads of nitrogen played an important role in the development of optimized scenarios to control nitrogen compounds with respect to acidification and eutrophication. First maps with critical loads of nitrogen were produced by the CCE in 1995 on the basis of national contributions, and, where not available, by applying the mass balance approach with European default values for forest sites. These maps were discussed by the Task Force on Mapping and adopted by the Working Group on Effects in 1995. The Task Force on Integrated Assessment Modelling (TFIAM) used the maps for the development of optimized nitrogen abatement scenarios for the ECE region. These scenarios formed the effect-oriented basis for the 1999 Gothenburg Protocol[6] concerning the reduction of emissions of sulphur dioxide (SO_2), nitrogen oxides (NO_x), volatile organic compounds (VOCs) and ammonia (NH_3).

The Gothenburg Protocol

The Gothenburg Protocol was amended in 2012 to include particulate matter (PM) and national emission reduction commitments for SO_2, NO_x, NH_3, VOCs and PM2.5 to be achieved in 2020. Again, the European critical load database compiled and updated by the CCE in collaborations with the National Focal Centres (Posch et al. 2011) was used to analyze the effects of emission reduction scenarios and thus supported the negotiations to revise the Protocol.

[5] www.unece.org/env/lrtap/taskforce/tfiam/welcome.html
[6] www.unece.org/env/lrtap/multi_h1.html

1.2 Mapping Critical Loads and Levels in Switzerland

Since 1990 Switzerland has participated in the programme for mapping critical loads and levels under the UNECE Convention on Long-range Transboundary Air Pollution. A National Focal Centre (NFC) was established at the Federal Office for the Environment[7]. The technical modelling and mapping tasks were carried out in cooperation with engineering companies and research institutes. The NFC was advised by scientists involved in various research fields such as air pollution, forests, soils, ecosystems or biodiversity.

Procedure for mapping critical loads in Switzerland

Critical loads data and maps used under the LRTAP Convention should cover the whole range of ecosystem sensitivity. Therefore, in Switzerland all ecosystems known to be sensitive towards acidifying deposition and towards eutrophying nitrogen deposition were included as far as feasible. According to the work programme under the Convention on LRTAP, high priority was given in 1991–1993 to mapping critical loads of acidity (sulfur and nitrogen). Forest soils and alpine lakes were selected as sensitive receptors (FOEFL 1994, Posch et al. 2007). In 1996, national maps of critical loads of nutrient nitrogen for forests and (semi-) natural ecosystems were produced and applied under the Convention (FOEFL 1996); they were based on the empirical method (UNECE 2016, Chapter V.2.1) and the so-called Simple Mass Balance (SMB) model (UNECE 2016, Chapter V.3.1). In addition to the SMB, multi-layer soil models for forest soils were applied in Switzerland to derive critical loads of acidity (Kurz et al. 1998).

Following the development of the scientific knowledge on the effects of nitrogen deposition and the corresponding mapping methods under the Convention, the Swiss critical loads of nutrient nitrogen were regularly updated and improved. For instance in 2007, the National Inventory of Dry Grasslands (Eggenberg et al. 2001, FOEN 2007) was included. The most recent update of the critical loads database was submitted to the CCE during the process of amending the Gothenburg Protocol (Achermann et al. 2011) and in 2015 as response to the data call 2014/15 of the Coordination Centre for Effects (Achermann et al. 2015, Slootweg et al. 2015).

In order to obtain information on the potential long-term ecological risks arising from exceedances of the critical loads of nitrogen, maps of present nitrogen depositions were regularly produced and compared with the critical load maps (FOEFL 1996, Kurz & Rihm 2001). The most recent critical load and exceedance maps for nutrient nitrogen are presented in detail in this volume. As in the past, the critical loads of nitrogen are based on the empirical method (chapter 2.2) and on the application of the Simple Mass Balance (SMB) model (chapter 2.3). The mapping of nitrogen deposition is described in chapter 3. Mapped critical loads of nitrogen and their exceedances are shown in chapter 4.

Besides the mapping of critical loads of acidity and nitrogen for forests and (semi-) natural ecosystems, further modelling, mapping or research activities were carried out

[7] www.bafu.admin.ch/luft/11640/11641/11644/index.html?lang=en

in the framework of International Cooperative Programmes under the Convention or at the national level addressing

- critical levels of ozone (e.g. Nussbaum et al. 2003, Waldner et al. 2007, Braun et al. 2014);
- critical loads of heavy metals (Achermann et al. 2005);
- critical levels of ammonia (Rihm et al. 2009, EKL 2014);
- impacts of air pollutants on materials (Reiss et al. 2004);
- dynamic modelling of air pollution effects on ecosystems due to acidification (Kurz et al. 1998a);
- dynamic modelling of eutrophying nitrogen deposition inducing changes in biodiversity (Sverdrup et al. 2008, Achermann et al. 2012, Achermann et al. 2014, Sverdrup & Belyazid 2015);
- the assessment of nitrogen deposition induced changes in biodiversity of selected (semi-)natural ecosystems along nitrogen deposition gradients (Roth et al. 2013, Roth et al. 2015); and
- the impacts of multi-year experimental nitrogen addition on sensitive subalpine grassland (Bassin et al. 2013).

2 > Methods to Derive Critical Loads of Nutrient Nitrogen

For estimating and mapping critical loads of nutrient nitrogen under the UNECE Convention on LRTAP two different approaches have been developed:

> Empirical critical loads: Based on published data on observed harmful effects of N deposition to sensitive (semi-)natural ecosystems, primarily in terms of changes in the structure and functioning of the ecosystems.
> Simple Mass Balance (SMB) method: Calculation of critical loads based on long-term nitrogen mass balance considerations for specific ecosystems.

The mapping manual (UNECE 2016) recommends both approaches and gives guidance to Parties to the Convention on how to use the approaches for the production of national critical load maps, from which the European maps can be derived at the CCE.

The following chapters give an overview of harmful effects of increased N deposition and present the application of the two methods in Switzerland. The empirical method was used to map the sensitivity of natural and semi-natural ecosystems and the SMB method was applied to productive forests.

2.1 Adverse Effects of Exposure to Atmospheric Nitrogen Compounds

The atmospheric nitrogen deposition in terrestrial and aquatic ecosystems in Switzerland as in all of Europe had increased substantially during the decades after 1950 (Schöpp et al. 2003). Since 1985 a decrease in air concentrations and depositions can be observed mainly for oxidized nitrogen compounds and to a lesser extent for reduced nitrogen compounds (BAFU 2016). Since 2000 the monitored concentrations of ammonia stay at the same level (Seitler & Thöni 2016). The main sources of reactive nitrogen in the atmosphere are emissions of NO_x from combustion processes and emissions of NH_3 from agricultural activities (FOEN 2016).

The impacts of increased nitrogen deposition and concentrations of gaseous nitrogen compounds (levels) upon biological systems are manifold, far-reaching and highly complex (Bobbink & Hettelingh 2011). The most important aspects are briefly summarized below.

2.1.1 Direct Toxicity of Nitrogen Gases

Sensitive plant species can be damaged by the direct exposure of above-ground parts of plants to gaseous pollutants in the air. In order to protect ecosystems from those direct effects **critical levels** have been defined as the "concentration, cumulative exposure or cumulative stomatal flux of atmospheric pollutants above which direct adverse effects on sensitive vegetation may occur according to present knowledge" (UNECE 2016). Critical levels were set for the nitrogen compounds nitrogen oxides (NO_x) and ammonia (NH_3) – besides sulphur dioxide (SO_2) and ozone (O_3).

Critical levels for nitrogen oxides

The critical level for the annual mean concentration of NO_x ($NO + NO_2$, expressed as NO_2) is set at 30 µg m^{-3} for all vegetation types (UNECE 2016). There is also a short-term critical level for NO_x (expressed as NO_2) defined as a 24-hour mean value of 75 µg m^{-3}.

Critical levels for ammonia

The critical level for the annual mean concentration of NH_3 is set at 3 µg m^{-3} (uncertainty range 2–4 µg m^{-3}) for higher plants, including heathland, grassland and forest ground flora. For lichens and bryophytes (including ecosystems where lichens and bryophytes are a key part of ecosystem integrity) the critical level is set at 1 µg m^{-3} (UNECE 2016). There is also a provisionally retained short-term critical level of ammonia (23 µg m^{-3}, monthly average) which is considered less reliable than the annual averages. Thus, for the purpose of mapping it is recommended to use only the annual mean values.

2.1.2 Eutrophication of (semi-)natural Ecosystems

Increased nitrogen deposition leads to N-accumulation in the soil and in the biomass. Depending on the stage of N saturation related to different soil types and ecosystem components, N loss from the ecosystem occurs by leaching to the groundwater (NO_3^-) and as gas (N_2, NO and N_2O, which is a greenhouse gas).

Nitrogen saturation of ecosystems

N saturation is defined as a condition where mineral N availability exceeds the absorption capacity of the ecosystem organisms. Signs of exceedances of this capacity are nitrate leaching with simultaneous losses of base cations or accumulation of ammonium in the soil. Nutritional imbalances due to nitrogen saturation, e.g. deficiencies of K, P, Mg, Ca, B, Mn relative to nitrogen are concluded to be a very important feature of the destabilization of sensitive ecosystems (e.g. forests).

Nitrogen deposition and biodiversity

N saturation and N excess also lead to changes in the competitive relationships between species, resulting in loss of biodiversity. Many (semi-)natural ecosystems are traditionally considered N-limited. The largest part of biodiversity is found in (semi-) natural ecosystems, both in aquatic and terrestrial habitats. Most of the plant species found in those habitats are adapted to nutrient-poor conditions and can only compete successfully on soils with low nitrogen availability. Under high nitrogen depositions many oligotrophic species will disappear or be replaced by nitrophilous species, which generally implies a loss of biodiversity and community uniqueness and a trend towards floristic homogenization. The loss of species richness in relation to elevated nitrogen

deposition also applies for mycorrhiza in forest soils that are important for the nutrient uptake by the fine roots of trees.

2.1.3 Soil Acidification

Contribution of nitrogen deposition to soil acidification

Soil acidification is defined as loss of acid neutralising capacity (ANC). Nitrogen deposition contributes to soil acidification due to nitrification processes and nitrate leaching. As a consequence, base cation leaching (Ca, K, Mg) is accelerated and the nutrient conditions for the plants change and can lead to nutritional imbalances. Low base cation-to-aluminium ratios affect the growth rates of many tree species and other plants. Plant species tolerating acidified conditions will gradually become dominant. The temporal evolution of the effects depends on changes in the buffering capacity of the soil, in base saturation and the chemical composition of the soil solution.

2.1.4 Increased Susceptibility to Secondary Stress Factors

High nitrogen deposition initially often leads to an ecosystem response in form of an increased growth rate and biomass production, followed by a destabilization phase. Ecosystems can be destabilized by a nitrogen induced sensitivity to secondary stress factors, e.g. lower stability of trees towards gales due to decreased root / shoot ratios and larger wood cells or increased susceptibility to frost, drought, insect pests and pathogens.

2.2 Empirical Critical Loads of Nitrogen

2.2.1 The Empirical Method

Empirical critical loads of nitrogen for (semi-)natural ecosystems are based on results of field studies, experiments and on scientific expert judgement. They are derived and updated in an established procedure as described by Bobbink & Hettelingh (2011, Chapter 2). The procedure includes the collection of scientific publications on nitrogen effects, drafting background documents for expert workshops, review of the background documents by international experts and setting critical load ranges for different ecosystem classes in the framework of scientific expert workshops held under the LRTAP Convention. The publications taken into consideration cover mainly long-term nitrogen addition field experiments and targeted field surveys, but also correlative or retrospective field observation studies along nitrogen deposition gradients.

International evaluation of empirical critical loads of nitrogen

The procedure for setting empirical critical loads was carried out the first time at the Lökeberg workshop (Grennfelt & Thörnelöf 1992). With the same procedure empirical critical loads were updated at a UNECE workshop held in Berne, Switzerland (Achermann & Bobbink 2003) and at a workshop held in Noordwijkerhout, The Netherlands, in 2010.

The outcome of the Noordwijkerhout workshop as presented in the workshop proceedings (Bobbink & Hettelingh 2011) was submitted to the Working Group on Effects of the Convention (UNECE 2010a). The Working Group invited National Focal Centres

Ecosystem types with empirical critical loads of nitrogen

(NFCs) to apply the revised empirical critical loads to national nature areas (UNECE 2010b). The empirical critical loads are listed in an overview table containing values for 47 different ecosystem types; additional information is given in footnotes. The table is arranged according to the EUNIS habitat codes[8]: (A) Marine habitats, (B) Coastal habitats, (C) Inland surface water habitats, (D) Mire, bog and fen habitats, (E) Grasslands and lands dominated by forbs, mosses and lichens, (F) Heathland, scrub and tundra habitats, (G) Woodland. Table 1 is an excerpt from the original overview table showing only those ecosystem types that are relevant for the Swiss critical load maps.

Tab. 1 > Overview of empirical critical loads of nitrogen (CL_{emp}[N]) for selected (semi-)natural ecosystem types

Units are in kg N ha^{-1} a^{-1}.

Ecosystem type	EUNIS code	CL_{emp}(N) range	Reliability[a]	Indication of critical load exceedance
[b] Coniferous woodland	G3	5–15	##	Changes in soil processes, nutrient imbalance, altered composition mycorrhiza and ground vegetation
[b] Broadleaved deciduous woodland	G1	10–20	##	Changes in soil processes, nutrient imbalance, altered composition mycorrhiza and ground vegetation
Arctic and (sub)- alpine scrub habitats	F2	5–15	#	Decline in lichens, bryophytes and evergreen shrubs
Sub-Atlantic semi-dry calcareous grassland	E1.26	15–25	##	Increase in tall grasses, decline in diversity, increased mineralization, N leaching; surface acidification
Molinia caerulea meadows	E3.51	15–25	(#)	Increase in tall graminoids, decreased diversity, decrease in bryophytes
Mountain hay meadows (see also chapter 2.2.2)	E2.3	10–20	(#)	Increase in nitrophilous graminoids, changes in diversity
(sub-)alpine grassland • acidic • calcareous	 E4.3 E4.4	 5–10 5–10	 # #	Changes in species composition; increase in plant production
[c] Permanent oligotrophic lakes / ponds	C1.1	3–10	##	Changes in species composition of macrophyte communities, increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
[d] Valley mires, poor fens and transition mires	D2	10–15	#	Increase in sedges and vascular plants, negative effects on bryophytes
[e] Rich fens	D4.1	15–30	(#)	Increase in tall graminoids, decrease in bryophytes
[f] Raised and blanket bogs	D1	5–10	##	Increase in vascular plants, altered growth and species composition of bryophytes, increased N in peat and peat water
[g] Alpine oligotrophic softwater lakes	C1.1	3–5	##	Phytoplankton community shift at N deposition 3–5; higher phytoplankton productivity at N deposition < 5

UNECE 2010a, Bobbink & Hettelingh 2011

a) The reliability is qualitatively indicated by ## reliable # quite reliable (#) expert judgement.

b) For application at broad geographical scales.

c) This critical load should only be applied to oligotrophic waters with low alkalinity with no significant agricultural or other human inputs. Apply the lower end of the range to boreal, sub-Arctic and alpine lakes, and the higher end of the range to Atlantic soft waters.

d) For EUNIS category D2.1 (valley mires): use the lower end of the range (#).

e) For high-latitude systems, apply the lower end of the range

f) Apply the high end of the range to areas with high levels of precipitation and the low end of the range to those with low precipitation levels; apply the low end of the range to systems with a low water table, and the high end of the range to those with a high water table. Note that water tables can be modified by management.

g) From ICP Waters (de Wit and Lindholm 2010).

8 http://eunis.eea.europa.eu/habitats-code-browser.jsp

For each ecosystem type the critical load is given as a range reflecting (1) the real intra-ecosystem variation within the ecosystem type, (2) the range of experimental treatments where an effect was observed or not observed and (3) uncertainties in estimated total atmospheric deposition values, where critical loads are based on field observations. The table also lists the indications of exceedance, i.e. the harmful changes in ecosystems to be expected when the critical load is exceeded in the long-term. The reliability of the given critical load values is indicated as follows:

- ## reliable: when several geographically separated studies showed similar results, or when experimental studies were supported by gradient studies
- # quite reliable: when one or two studies supported the range given;
- (#) expert judgement: when results from experimental studies were lacking for this type of ecosystem. The critical load is then based upon knowledge of ecosystems which are likely to be comparable with this ecosystem.

In addition, the International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes (ICP Waters) has carried out a broad literature review on nutrient enrichment effects of atmospheric N deposition on biology in oligotrophic surface waters (de Wit and Lindholm 2010). As a result of this evaluation they propose an additional empirical critical load of nitrogen of 3–5 kg N ha^{-1} a^{-1} for oligotrophic softwater alpine and boreal lakes.

2.2.2 Derivation of Country-specific Empirical Critical Loads

For deriving exposure-response relationships for vascular plants in Swiss mountainous ecosystems, nitrogen deposition at high spatial resolution (100 x 100 m^2 grid) and site-specific plant occurrence data from the Swiss Biodiversity Monitoring (BDM[9]) were analysed. The analysis has led to quantitative exposure-response relationships for species-rich mountain hay meadows (EUNIS class E2.3, Roth et al. 2013) and for alpine and subalpine scrub habitats (EUNIS class F2.2, Achermann et al. 2014). It was tested (i) whether N deposition is negatively related to plant species richness and (ii) whether N deposition is related to species richness of oligotrophic species.

In addition to the explanatory variable “N deposition” further environmental factors being known to influence plant diversity from earlier studies were taken into account (Wohlgemuth et al. 2008, Roth et al. 2013). They include altitude, inclination, exposition, precipitation and indicator values for soil pH and soil humidity.

Mountain hay meadows

In mountain hay meadows mean ±SD species richness was 38.3 ± 7.45, from which 9.58 ± 9.71 oligotrophic species. Total species richness and species richness of oligotrophic species were negatively related to N deposition. After adjusting for confounding effects of the environmental factors, the exposure-response relationship between N deposition and total species richness was $58.935e^{-0.018x}$, $R^2 = 0.55$ (Figure 2); between N deposition and species richness of oligotrophic species it was $51.186e^{-0.114x}$, $R^2 = 0.71$ (Figure 3). As expected, the response of oligotrophic species to N deposition was more pronounced than the response of overall species richness.

[9] www.biodiversitymonitoring.ch

Fig. 2 > Predicted species richness in mountain hay meadows as a function of N deposition, after adjusting for effects of additional environmental factors

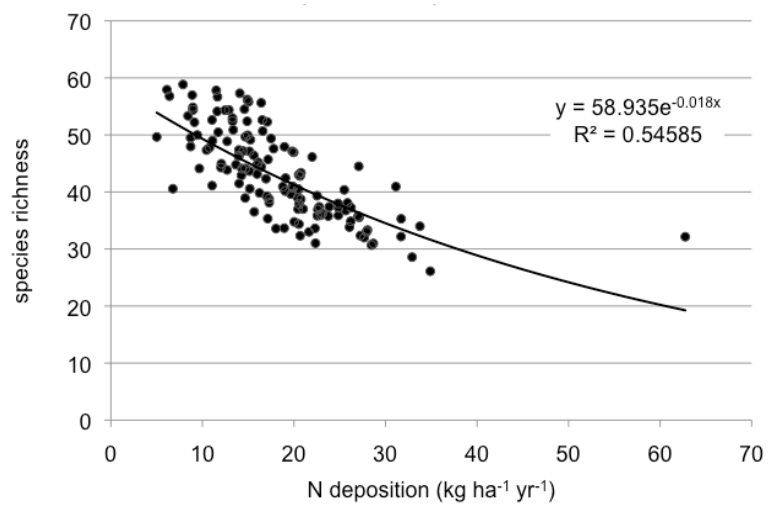
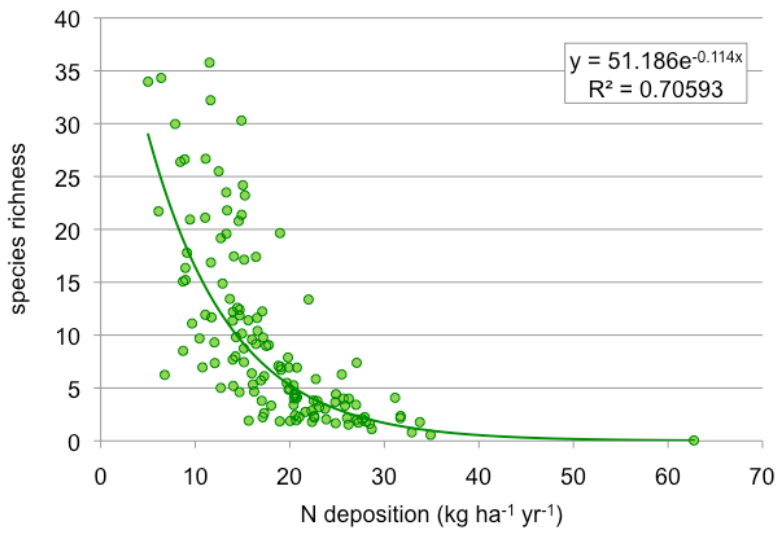


Fig. 3 > Predicted species richness of oligotrophic species in mountain hay meadows as a function of N deposition, after adjusting for effects of additional environmental factors



Alpine and subalpine scrub habitats

In alpine and subalpine scrub habitats the mean ±SD species richness was 28.05 ± 12.29, from which 22.19 ± 9.26 oligotrophic species. Species richness and species richness of oligotrophic species were slightly negatively related to N deposition.

After adjusting for confounding effects of the environmental factors, the exposure-response relationship between nitrogen deposition and total species richness was $43.69e^{-0.05x}$, $R^2 = 0.27$ (Figure 4); between nitrogen deposition and species richness of oligotrophic species it was $43.75e^{-0.076x}$, $R^2 = 0.42$ (Figure 5). As expected, the response of oligotrophic species to N deposition was slightly more pronounced than the response of overall species richness.

Fig. 4 > Predicted species richness in alpine and subalpine scrub habitat (F2.2) as a function of N deposition, after adjusting for effects of additional environmental factors

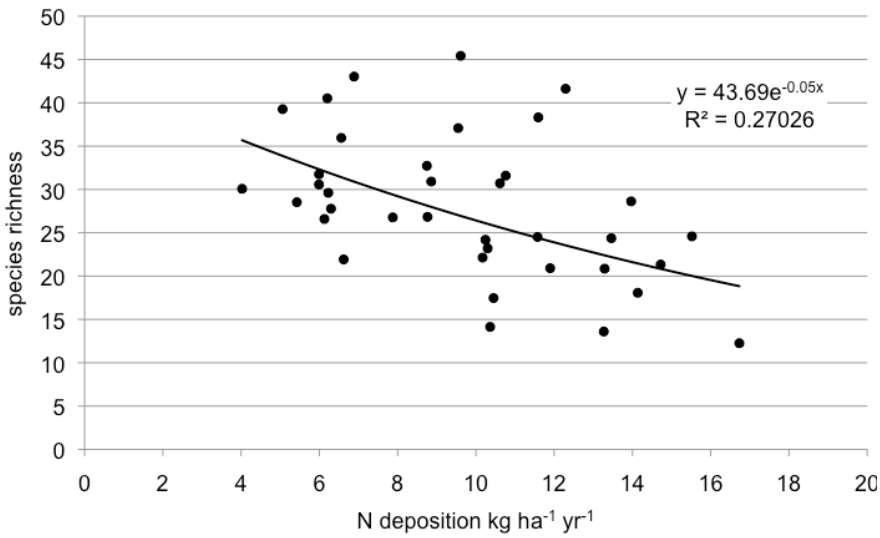
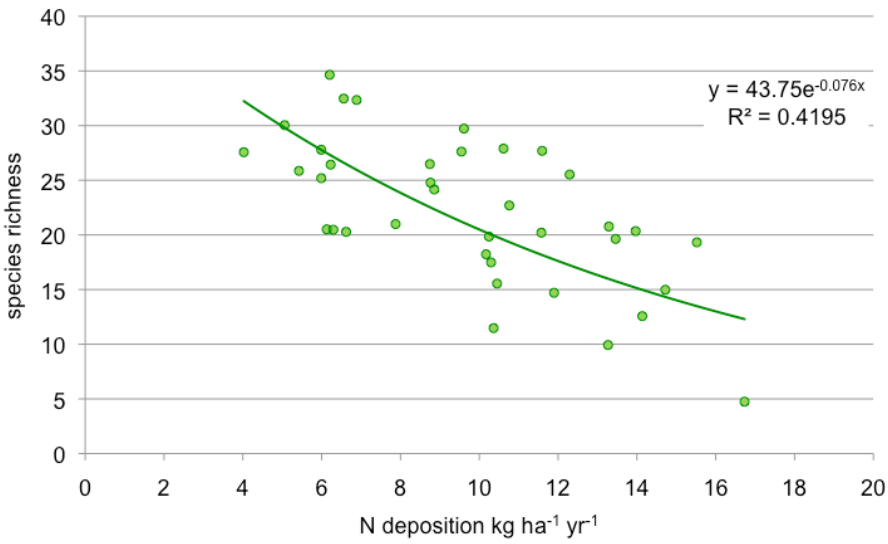


Fig. 5 > Species richness of oligotrophic species in alpine and subalpine scrub habitat (F2.2) as a function of N deposition, after adjusting for effects of additional environmental factors



Change point models to estimate critical loads

If change point models are used to estimate empirical critical loads of nitrogen for mountain hay meadows and for (sub-)alpine scrub habitats using the above mentioned data sets, it could be concluded that the range of the empirical critical load set for mountain hay meadows (10–20 kg N ha^{-1} a^{-1}) could be narrowed down to 10–15 kg N ha^{-1} a^{-1}, whilst the empirical critical load set for (sub-)alpine scrub habitats (5–15 kg N ha^{-1} a^{-1}) was confirmed (Roth et al. 2017).

In alpine and subalpine scrub habitats (F2.2) the exposure-response relationships for total species richness and for species richness of oligotrophic species are very similar (Figure 4, Figure 5). This is not surprising, since on average 79 ± 13% of all species belong to the oligotrophic species. The effect of relatively small doses, especially on oligotrophic species, is in accordance with recent studies in other alpine habitats, where the proportional biomass of functional groups changed by the addition of 5 kg N $ha^{-1}a^{-1}$ (Bassin et al. 2013).

2.2.3 Mapping Empirical Critical Loads for Ecosystems in Switzerland

For the purpose of applying the empirical critical loads approach, countries have to identify those receptors or ecosystems of maximum sensitivity from the list of ecosystem types of Table 1 relating to their individual priorities to protect the environment. Table 2 and Table 3 present the ecosystems for which the empirical critical load approach was applied in Switzerland. Table 3 specifically addresses dry grassland (TWW).

Selection of sensitive ecosystems

The selection of sensitive ecosystems to be protected by applying the empirical method is based on ecosystem and vegetation data compiled from various sources described below. All of the selected ecosystems are of high conservation importance with respect to biodiversity, landscape quality and ecosystem services. They include natural as well as semi-natural ecosystem types. Overall, 45 sensitive ecosystem types according to EUNIS classes were identified and included in the critical load data set:

- 21 various so-called “types of vegetation worthy of protection” from the vegetation atlas by Hegg et al. (1993) including rare and species-rich forest types, alpine heaths, grasslands and surface waters (see Tab. 2). The atlas contains distribution maps for 97 vegetation types with a resolution of 1 x 1 km². 21 vegetation types sensitive to eutrophication were selected.
- 1 type of mountain hay meadow (see Tab. 2) in montane to sub-alpine altitudinal zones with more than 35 species per 10 m² (Roth et al. 2013). This applies to 122 sites of the Swiss Biodiversity Monitoring (BDM, indicator Z9[10]).
- 1 type of raised bog from the Federal Inventory of Raised and Transitional Bogs of National Importance (Appendix to Swiss Confederation 1991) (see Tab. 2). This data set is available in vector format at a scale of 1:25 000. The inventory contains only bogs with relevant occurrences of *sphagnion fusci*.
- 3 types of poor or rich fens from the Federal Inventory of Fenlands of National Importance (Appendix to Swiss Confederation 1994, WSL 1993) (see Tab. 2). This data set is available in vector format at a scale of 1:25 000. For the maps compiled by the Coordination Centre for Effects (UNECE), only the mesotrophic fens were selected. Eutrophic fens, such as *Phragmition*, *Magnocaricon*, *Molinion* and *Calthion* were omitted.
- 1 type of oligotrophic alpine lakes (see Tab. 2). The catchments of 100 lakes in Southern Switzerland at altitudes between 1650 and 2700 m (average 2200 m) were mapped by Posch et al. 2007. To a large extent the selected catchments consist of crystalline bedrock and are therefore sensitive to acidification and eutrophication.

[10] www.biodiversitymonitoring.ch/en/data/indicators/z/z9.html

> 18 types of dry grassland (TWW) from the National Inventory of Dry Grasslands of National Importance (Eggenberg et al. 2001, FOEN 2007) (see Tab. 3). This data set is available in vector format at a scale of 1:25 000. Many of those grasslands are extensively managed as hay meadows. They also include alpine and subalpine grassland.

For most of the selected ecosystems a value in the middle of the proposed range was chosen as the critical load. Only in the case of fens and several grassland types the critical load value was set near the lower end of the range because the respective ecosystems are considered to be more sensitive than the average, since they are nutrient poor and located in relatively high altitudes with short vegetation periods.

Tab. 2 > Selected ecosystems for the application of empirical critical loads, $CL_{emp}(N)$, applied in Switzerland

Ecosystem type	$CL_{emp}(N)$ range	Relevant vegetation types in Switzerland	EUNIS code	$CL_{emp}(N)$
Coniferous forests (EUNIS G3)	5–15	Molinio-Pinetum *(Pfeifengras-Föhrenwald)* Ononido-Pinion *(Hauhechel-Föhrenwald)* Cytiso-Pinion *(Geissklee-Föhrenwald)* Calluno-Pinetum *(Heidekraut-Föhrenwald)* Erico-Pinion mugi (Ca) *(Erika-Bergföhrenwald auf Kalk)* Erico-Pinion sylvestris *(Erika-Föhrenwald)*	G3.44 G3.43 G3.4 G3.3 G3.44 G3.44	12 12 12 10 12 12
Deciduous forests (EUNIS G1)	10–20	Quercion robori-petraeae *(Traubeneichenwald)* Quercion pubescentis *(Flaumeichenwald)* Fraxino orno-Ostryon *(Mannaeschen-Hopfenbuchwald)*	G1.7 G1.71 G1.73	15 15 15
Arctic and (sub)-alpine scrub habitats (EUNIS F2)	5–15	Juniperion nanae *(Zwergwacholderheiden)* Loiseleurio-Vaccinion *(Alpenazaleenheiden)*	F2.23 F2.21	10 10
Sub-atlantic semi-dry calcareous grassland	15–25	Mesobromion (erecti) *(Trespen-Halbtrockenrasen)*	E1.26	15
Molinia caerulea meadows	15–25	Molinion (caeruleae) *(Pfeifengrasrieder)*	E3.51	15
Mountain hay meadows[a]	10–15	Grassland types 4.5.1–4.5.4 (Delarze et al. 2008)	E2.3	12
(sub)-alpine grassland (EUNIS E4)	5–10	Chrysopogonetum grylli *(Goldbart-Halbtrockenrasen)* Seslerio-Bromion (Koelerio-Seslerion) *(Blaugras-Trespen-Halbtrockenrasen)* Stipo-Poion molinerii *(Engadiner Steppenrasen)*, sub-alpine Elynion *(Nacktriedrasen)*, alpine	E4.3 E4.4 E4.4 E4.42	10 10 10 7
Poor fens (EUNIS D2)	10–15	Scheuchzerietalia *(Scheuchzergras)* Caricion fuscae *(Braunseggenried)*	D2.21 D2.2	10 12
Rich fens	15–30	Caricion davallianae *(Davallsseggenried)*	D4.1	15
Raised bogs	5–10	Sphagnion fusci *(Hochmoor)*	D1.1	7
Permanent oligotrophic lakes / ponds	3–10	Littorellion *(Strandling-Gesellschaften)*	C1.1	7
Alpine oligotrophic softwater lakes	3–5	sensitive alpine lakes in Southern Switzerland	C1.1	4

[a] adapted according to Roth et al. 2017

Units are in kg N ha^{-1} a^{-1}

Tab. 3 > Empirical critical loads of nitrogen, $CL_{emp}(N)$, assigned to dry grasslands (TWW) of the National Inventory of Dry Grasslands

In kg N ha^{-1} a^{-1}.

TWW-code		Vegetation type	EUNIS	Remarks	$CL_{emp}(N)$
1	CA	Caricion austro-alpinae *(Südalpine Blaugrashalde)*	E4.4	(sub-)alpine grassland	8
2	CB	Cirsio-Brachypodion *(Subkontinentaler Trockenrasen)*	E1.23	similar to TWW 18 (Mesobromion), also used as hay meadow	12
3	FP	Festucion paniculatae *(Goldschwingelhalde)*	E4.3	similar to TWW 13 (Festucion variae); also mapped by Hegg et al.	7
4	LL	low diversity, low altitude grassland *(artenarme Trockenrasen der tieferen Lagen)*	E2.2	contains different types, promising diversity when mown, therefore lower range chosen	15
5	AI	Agropyrion intermedia *(Halbruderaler Trockenrasen)*	E1.2	transitional type	15
6	SP	Stipo-Poion *(Steppenartiger Trockenrasen)*	E1.24	pastures/fallows in large inner-alpine valleys; $CL_{emp}(N)$ based on national expert-judgment (Hegg et al. 1993)	10
7	MB_{SP}	Mesobromion / Stipo-Poion *(Steppenartiger Halbtrockenrasen)*	E1.26	pastures, slightly more nutrient-rich than Mesobromion (TWW18)	15
8	XB	Xerobromion *(Subatlantischer Trockenrasen)*	E1.27	meadows/pastures/fallows in large inner-alpine valleys; $CL_{emp}(N)$ based on national expert-judgment (Hegg et al. 1993)	12
9	MB_{XB}	Mesobromion/Xerobromion *(Trockener Halbrockenrasen)*	E1.26	similar to TWW 18 (Mesobromion)	12
10	LH	low diversity, high altitude grassland *(artenarme Trockenrasen der höheren Lagen)*	E2.3	contains different types of dry grassland at high altitude	12
11	CF	Caricion ferrugineae *(Rostseggenhalde)*	E4.41	(sub-)alpine grassland; also mapped by Hegg et al.	7
12	AE	Arrhenatherion elatioris *(Trockene artenreiche Fettwiese)*	E2.2	often used as meadows, lower range chosen as it occurs at all altitude levels	12
13	FV	Festucion variae *(Buntschwingelhalde)*	E4.3	(sub-)alpine grassland, middle of the range chosen	7
14	SV	Seslerion variae *(Blaugrashalde)*	E4.43	alpine grassland, middle of the range chosen; also mapped by Hegg et al.	7
15	NS	Nardion strictae *(Borstgrasrasen)*	E1.71	meadows, subalpine	12
16	OR	Origanietalia *(Trockene Saumgesellschaft)*	E2.3	meadows/fallows	15
17	MB_{AE}	Mesobromion/Arrhenatherion *(Nährstoffreicher Halbtrockenrasen)*	E1.26	slightly more nutrient-rich than Mesobromion (TWW18)	15
18	MB	Mesobromion *(Echter Halbrockenrasen)*	E1.26	genuine semi-dry grassland	12

Eggenberg et al. 2001, FOEN 2007

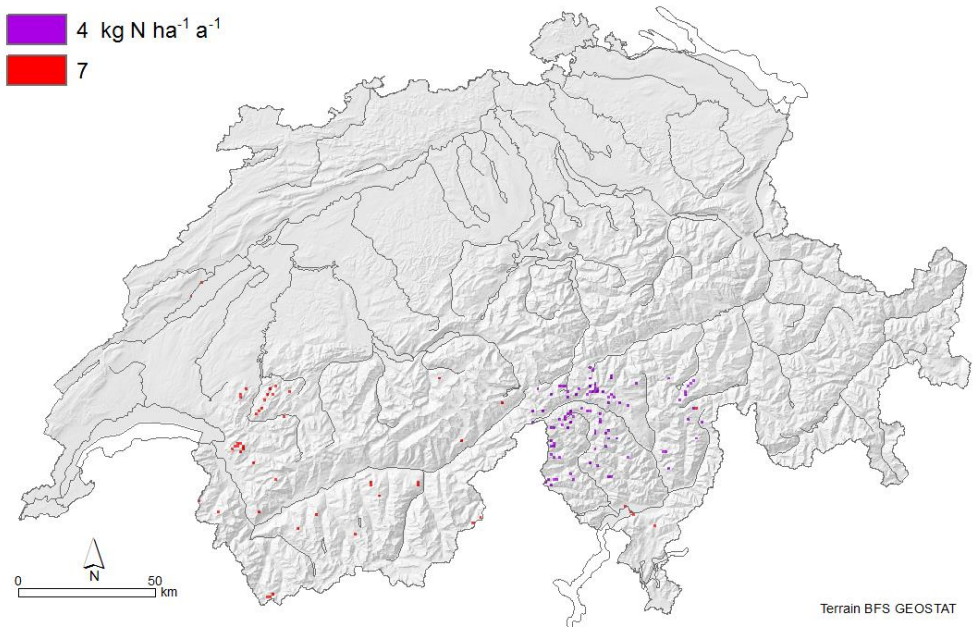
Raised bogs, oligotrophic ponds, alpine grassland, alpine heaths and most of the selected forest types are (semi-)natural ecosystems, i.e. they are not managed or only poorly managed.

Fens and species-rich grassland below the alpine level are semi-natural systems, in general. They developed under permanent traditional management over centuries. When these extensive forms of management change, the ecosystems generally show a decrease in biodiversity.

The TWW data set complements well the grassland types mapped by Hegg et al. (1993). It contains 18 vegetation groups, which partially also occur in the inventory of Hegg. The two inventories are used here in a complementary way, because they fulfil different purposes: The atlas by Hegg gives an overview of the occurrence of selected vegetation types, while TWW focuses on the precise description of sites with national importance.

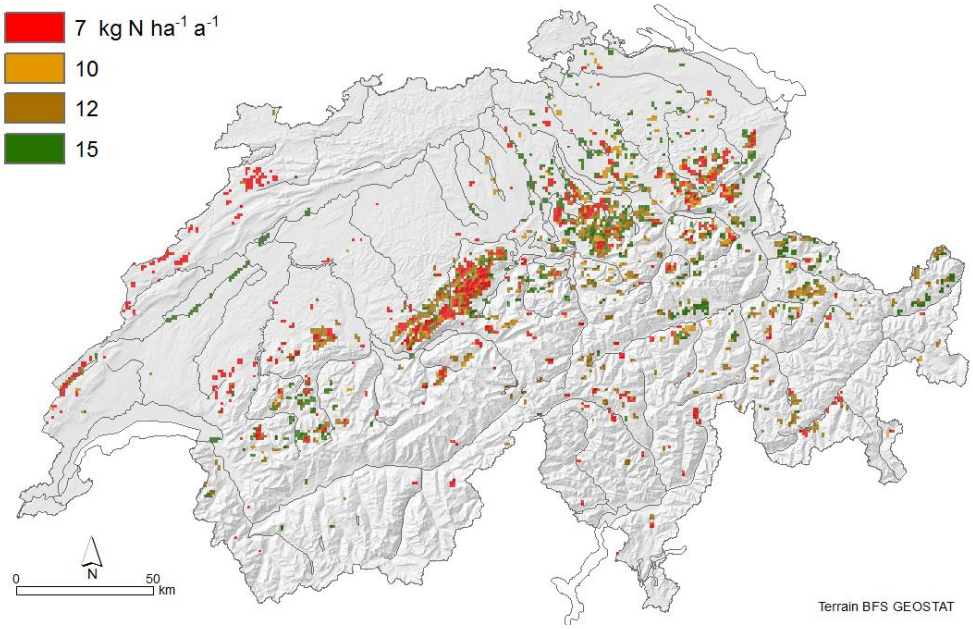
For the data and maps used under the Convention on Long-range Transboundary Air Pollution and submitted to the Coordination Centre for Effects, the outlines of all ecosystem-specific polygons (in vector format) were converted to a 1 x 1 km² grid (present / absent criterion). If more than one ecosystem type occurs within a 1 x 1 km² grid-cell, the lowest value of $CL_{emp}(N)$ was selected for this cell. The spatial distribution of the selected ecosystem types and their $CL_{emp}(N)$ according to Table 2 and Table 3 are shown on the maps in Figure 6 to Figure 10.

Fig. 6 > $CL_{emp}(N)$ for oligotrophic surface waters (EUNIS code C)



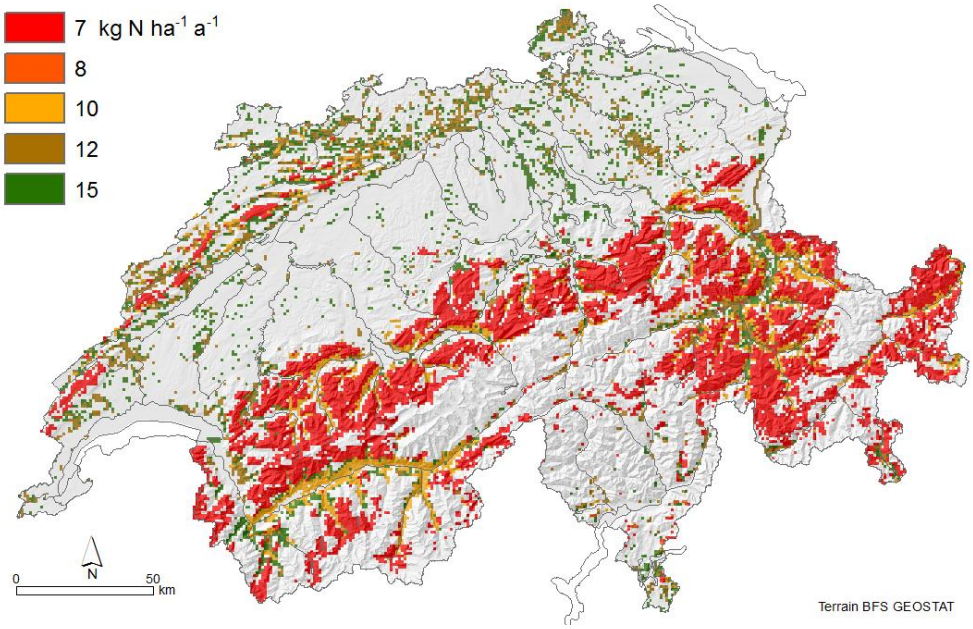
Hegg et al. 1993, Posch et al. 2007

Fig. 7 > $CL_{emp}(N)$ for raised bogs and fens (EUNIS code D)



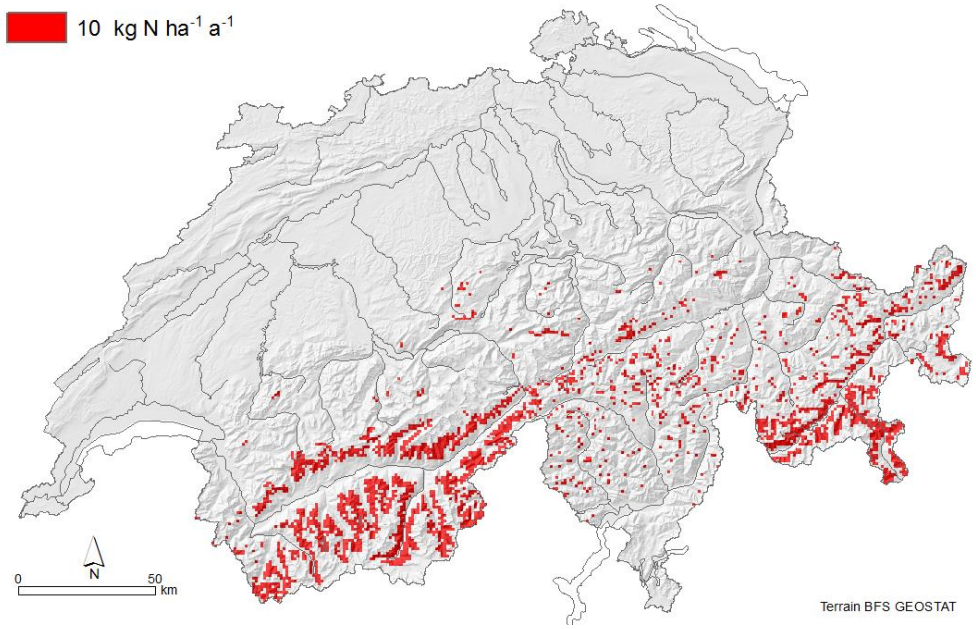
Swiss Confederation 1991 and 1994

Fig. 8 > $CL_{emp}(N)$ for selected grassland types (EUNIS code E)



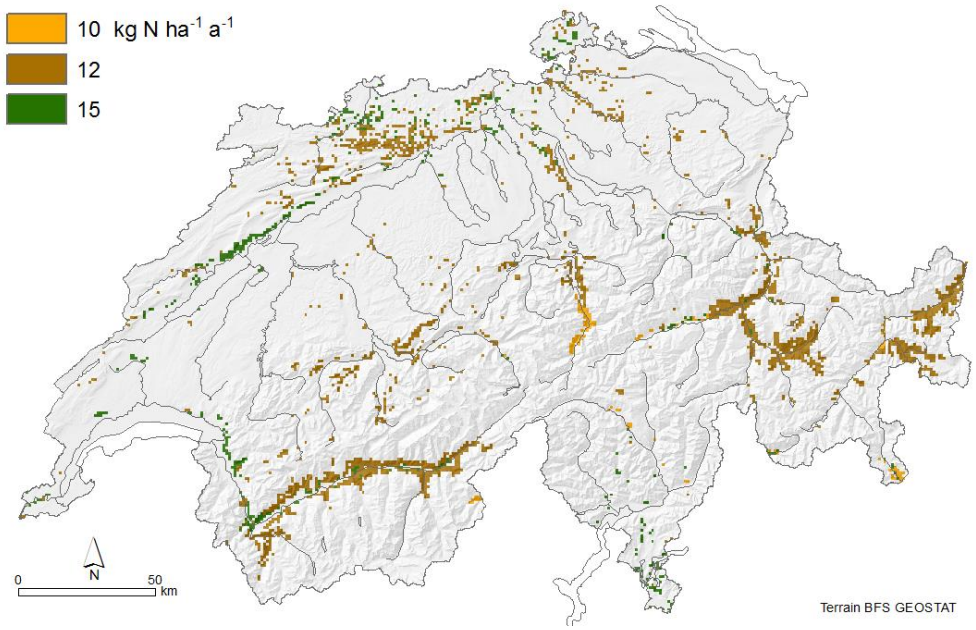
Hegg et al. 1993, TWW, BDM

Fig. 9 > $CL_{emp}(N)$ for selected alpine scrub habitats (EUNIS code F)



Hegg et al. 1993

Fig. 10 > $CL_{emp}(N)$ for selected forest types (EUNIS code G)



Hegg et al. 1993

2.3 The Simple Mass Balance Method (SMB)

In Switzerland, the SMB method was applied for productive forest ecosystems, i.e. sites where wood harvesting is possible.

2.3.1 The Calculation Method

The SMB method for calculating critical loads of nutrient nitrogen is a steady-state model based on the nitrogen saturation concept. There are several definitions of N saturation. In this context it is understood as follows: The atmospheric nitrogen deposition must not lead to a situation where the availability of inorganic nitrogen is in excess of the total combined plant and microbial nutritional demand. I.e. the inputs should not be larger than the natural sinks plus outputs.

Principle of the Simple Mass Balance (SMB)

The basic principle of the SMB method is to identify the long-term average sources and sinks of inorganic nitrogen in the system, and to determine the maximum tolerable nitrogen input that will protect the system from nitrogen saturation. The nitrogen cycling within the ecosystems is mainly regulated by biological processes that depend on the following factors: (1) the ecosystem type, (2) former and present land use and management, (3) environmental conditions, especially those influencing the nitrification rate and the immobilization rate. Consequently these factors are important for calculating and setting critical loads of nutrient nitrogen.

In the mapping manual (UNECE 2016) a simplified SMB equation is formulated as follows:

$$CL_{nut}(N) = N_i + N_u + N_{de} + N_{le(acc)} \quad (2.1)$$

Where:

$CL_{nut}(N)$ *critical load of nutrient nitrogen [kg N ha^{-1} a^{-1}].*

N_i = *acceptable immobilization rate of N in soil organic matter (including forest floor) at N inputs equal to critical load, at which adverse ecosystem change will not take place [kg N ha^{-1} a^{-1}].*

N_u *nitrogen uptake; net removal of nitrogen in vegetation at critical load [kg N ha^{-1} a^{-1}]. This is the amount of nitrogen which is removed from the system by (wood) harvesting.*

N_{de} *denitrification rate [kg N ha^{-1} a^{-1}]. This is the flux to the atmosphere of gaseous compounds (N_2, N_2O and NO) produced by microorganisms (mainly under anaerobic conditions) in the soil.*

$N_{le(acc)}$ *acceptable total nitrogen leaching from the rooting zone at which no damage occurs in the terrestrial or linked ecosystem plus any enhanced leaching following forest harvesting [kg N ha^{-1} a^{-1}]. This is the N removed from the soil by the vertically percolating water flux.*

The equation is based on the following assumptions:

- All rates and fluxes of the involved processes are represented by annual means.
- Temporal variations in N_u as a function of forest age and management are not included, i.e. the temporal scale of the investigation is longer than one rotation period (>100 years).
- Nitrogen losses by natural fires, erosion and ammonia volatilisation as well as biological N fixation are negligible in most Swiss forests.

Equation 2.1 as such is not operational, as N_{de} strongly depends on nitrogen deposition. The mapping manual proposes to use linear or non-linear functions to calculate N_{de}. For the application in Switzerland the constant function is used:

2.2

$$N_{de} = f_{de} \cdot (N_{dep} - N_u - N_i)$$

$$\text{if } N_{dep} > N_u + N_i$$

$$N_{de} = 0 \quad \text{otherwise}$$

Where:

f_{de} *denitrification fraction.*
N_{dep} *nitrogen deposition [kg N ha^{-1} a^{-1}].*

This formulation implicitly assumes that immobilization and uptake are faster processes than denitrification. Under critical load conditions N_{dep} is equal to $CL_{nut}(N)$. By inserting equation 2.2 into 2.1 the critical load becomes:

2.3

$$CL_{nut}(N) = N_i + N_u + N_{le(acc)} / (1 - f_{de})$$

Maps of critical loads to be used under the Convention on LRTAP were produced in two steps:

Application of the SMB method for forest sites

In a first step, equation 2.3 was applied for 10 331 sites of the National Forest Inventory NFI (WSL 1990/92, EAFV 1988) located on a 1 x 1 km² raster, and for 301 forest sites used in dynamic modelling (Achermann et al. 2015). Thereby, only NFI-sites with a defined mixing ratio of deciduous and coniferous trees are included. This corresponds approximately to the productive forest area as unproductive and unmanaged woodlands such as brush forests and inaccessible forests are excluded.

In a second step, the lower limit of $CL_{nut}(N)$ calculated by the SMB was set to 10 kg N ha^{-1} a^{-1} (corresponding to the lower limit of $CL_{emp}(N)$ used for forests). This means, all values of $CL_{nut}(N)$ below 10 kg N ha^{-1} a^{-1} were set to 10. This was done with respect to the fact that so far no empirically observed harmful effects in forest ecosystems were published for depositions lower than 10 kg N ha^{-1} a^{-1} and for latitudes and altitudes

typical for Switzerland. Therefore, the critical loads calculated with the SMB method were adjusted to empirically confirmed values.

2.3.2 Quantification of the Input Parameters

Table 4 gives a summary of the input parameter values applied for the national maps.

Tab. 4 > Ranges of input parameters used for calculating $CL_{nut}(N)$ with the SMB method

Parameter	Values	Comment
$N_{le(acc)}$	4 kg N ha^{-1} a^{-1} at 500 m a.s.l., 2 kg N ha^{-1} a^{-1} at 2000 m a.s.l., linear interpolation in-between	Acceptable N leaching. Leaching mainly occurs by management (after cutting), which is more intense at lower altitudes.
N_i	1.5 kg N ha^{-1} a^{-1} at 500 m a.s.l., 2.5 kg N ha^{-1} a^{-1} at 1500 m a.s.l., linear interpolation in-between	N immobilization in the soil. At low temperature (correlated with high altitude) the decomposition of organic matter slows down and therefore the accumulation rates of N are naturally higher.
N_u	0.5–14.7 kg N ha^{-1} a^{-1}	N uptake calculated on the basis of long-term harvesting rates in managed forest ecosystems.
f_{de}	0.2–0.7 depending on the wetness of the soil	Denitrification fraction. For sites of the National Forest Inventory (NFI), information on wetness originates from soil map 1:200 000. For sites with application of dynamic models (DM-sites) it is a classification according to the depth of the water logged horizon.

Long-term sustainable nitrogen immobilization

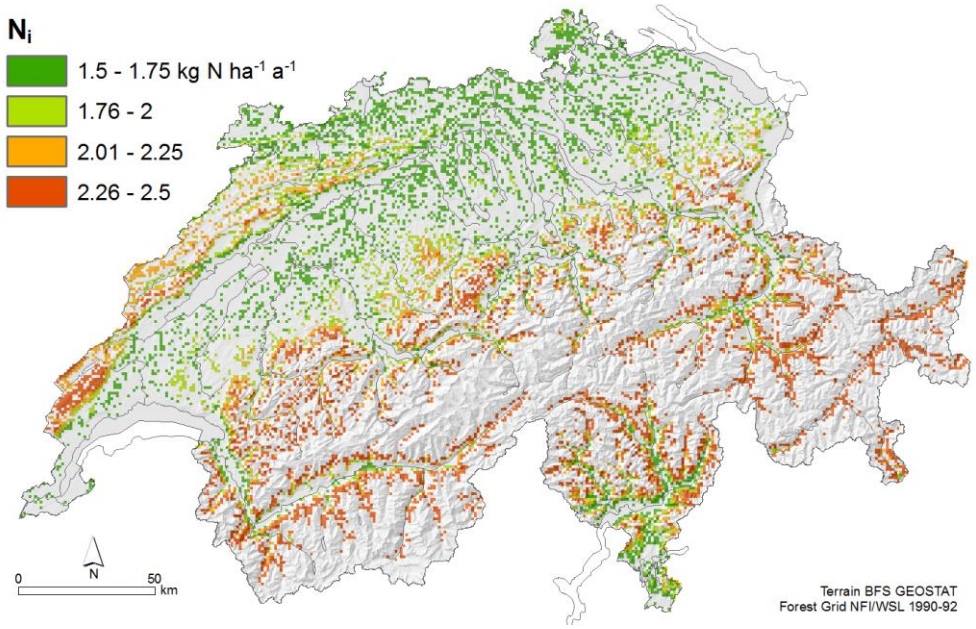
a) Nitrogen immobilization

According to the mapping manual (UNECE 2016) there is no full consensus yet on long-term sustainable immobilization rates. E.g. in Swedish forest soils the annual N immobilisation since the last glaciation was estimated at 0.2–0.5 kg N ha^{-1} a^{-1} (Rosén et al. 1992). Considering that immobilization of N is probably higher in warmer climates, the mapping manual states that values up to 1 kg N ha^{-1} a^{-1} could be used without causing unsustainable accumulation of N in the soil and that even higher values were used for critical load calculations. Considering these uncertainties the following (relatively high) values are used for the Swiss critical load map:

$N_i = 1.5$ *kg N* ha^{-1} a^{-1} *at low altitudes (<500 m a.s.l.)*
$N_i = 2.5$ *kg N* ha^{-1} a^{-1} *at high altitudes (>1500 m a.s.l.)*

At altitudes in-between, N_i is calculated by linear interpolation (see Figure 11). The reason for the altitude dependency is the retarded decomposition of organic matter at lower temperatures and/or higher precipitation; therefore the accumulation rates of soil organic matter (including N and carbon) are naturally higher at high altitudes. The increase of the carbon content in the soil with altitude for large parts of Switzerland was shown on recent maps by Nussbaum et al. (2012).

Fig. 11 > Nitrogen immobilization values for forest soils used for the SMB method on the 1 x 1 km² raster



b) Nitrogen uptake

Nitrogen uptake

Nitrogen and base cations (Ca, Mg, K, Na) are taken up by trees and used for biomass production. In unmanaged forests at steady state, the net uptake will be zero since biomass production is in balance with biomass decomposition. In a managed forest, the net uptake rate is obtained by multiplying the long-term net growth (harvesting) rate with nitrogen and cation contents of the wood.

For the 301 sites used in dynamic modelling (DM-sites), Kurz & Posch (2015) modelled net-uptake fluxes with MakeDep (Alveteg et al. 2002) using biomass data from the third National Forest Inventory (NFI, WSL 2013), tree genera-specific logistic growth curves, site productivity index, nutrient contents in the various compartments of the tree, and average annual harvesting rates (FOEN 2013). MakeDep is able to consider the mutual dependence of deposition, forest canopy growth/size and nutrient demand of the growing forest.

Harvesting rates were stratified according to the five NFI-regions: Jura, Central Plateau, Pre-Alps, Alps and Southern Alps. The harvesting rates before 2000 add up to 4.5 million m³ stem wood, which is in the range of the typical annual harvest in Switzerland (in years without heavy storms); after 2000, the harvest of energy wood slightly increased and the average harvest was around 5.0 million m³ (FOEN 2013).

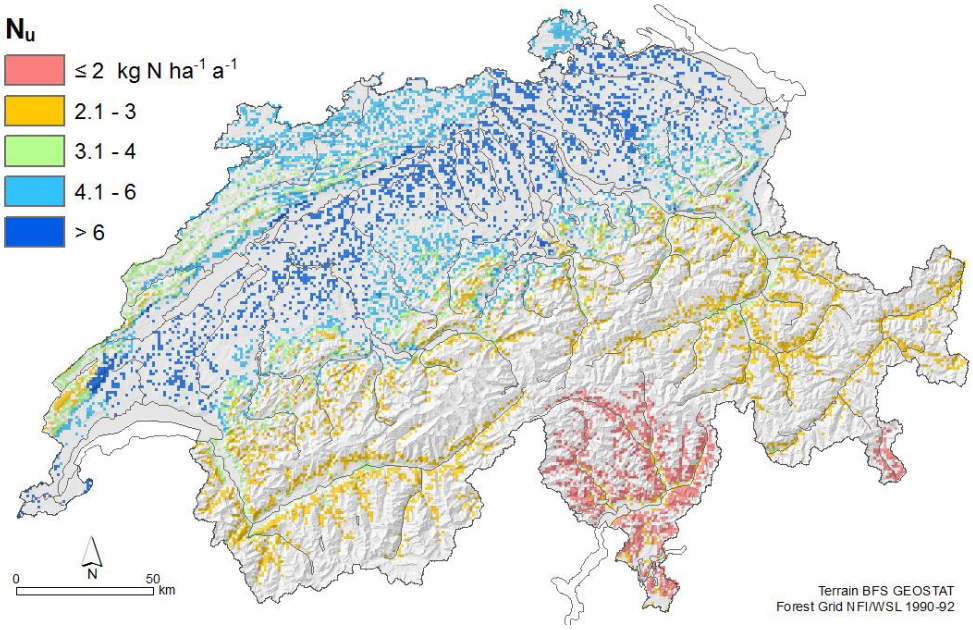
The nitrogen uptake for the forest sites on the 1 x 1 km² raster was derived from the results at the DM-sites by linear regressions with altitude (z); the regression analysis was stratified according to the five NFI-regions (Table 5, Figure 12). In the Jura and Central Plateau regions, the average harvesting rates reach the long-term gross growth, but in the mountainous and southern parts of Switzerland it is much lower than gross

growth. The resulting uptake values are in the range from 1.0 to 8.8 kg N $ha^{-1}a^{-1}$ (see Figure 12).

Tab. 5 > Net nitrogen uptake (N_u) in the five NFI-regions (kg N ha^{-1} a^{-1})

Region	Average	Function of altitude z (m a.s.l.)
1. Jura	5.3	6.99–0.00300 z
2. Central Plateau	8.5	8.5
3. Pre-Alps	4.3	7.60–0.00322 z
4. Alps	2.9	3.58–0.00064 z
5. Southern Alps	1.6	2.29–0.00056 z
Average CH	4.4	-

Fig. 12 > Net nitrogen uptake (N_u) of managed forest ecosystems used for the SMB method on the 1 x 1 km² raster



c) Nitrogen leaching

Acceptable nitrogen leaching

Within the scope of the CCE data-call 2007, the National Focal Centres were requested to reassess their $CL_{nut}(N)$ calculations and update them if appropriate based on revised critical N concentrations (cNacc) (UNECE 2013, chapter 5.3.1.2). For Switzerland, the proposed values for cNacc were tested (see Achermann et al. 2007). Some of the proposed values led to implausible high N leaching and $CL_{nut}(N)$, mainly in high precipitation areas.

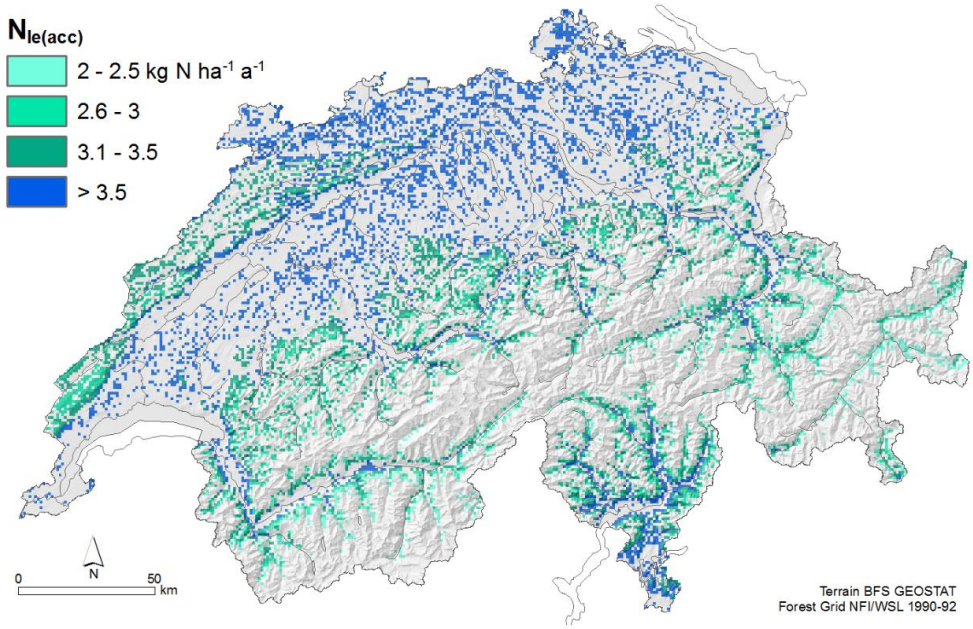
Therefore it was decided to continue using the acceptable N leaching rates ($N_{le[acc]}$), which were used already in former data submissions. They are basically drawn from earlier versions of the Mapping Manual (UNECE 1996) and on findings of the work-

shop on critical loads for nitrogen held in Lökeberg in 1992 (Grennfelt and Thörnelöf 1992):

$$N_{le(acc)} = 4 \text{ kg N ha}^{-1} \text{ a}^{-1} \text{ at low altitudes (<500 m a.s.l.)}$$
$$N_{le(acc)} = 2 \text{ kg N ha}^{-1} \text{ a}^{-1} \text{ at high altitudes (>2000 m a.s.l.)}$$

At altitudes in-between, $N_{le(acc)}$ is calculated by linear interpolation. The rationale for this procedure is that acceptable leaching mainly occurs after disturbances by management (cutting), which is more intense at lower altitudes. The resulting map is shown in Figure 13.

Fig. 13 > Acceptable nitrogen leaching values of forest ecosystems used for the SMB method on the 1 x 1 km² raster



d) Denitrification fraction

Denitrification

The f_{de} values proposed in the mapping manual (UNECE 2016) are between 0.0 and 0.8. There are two proposals for relating f_{de} values to different soil properties:

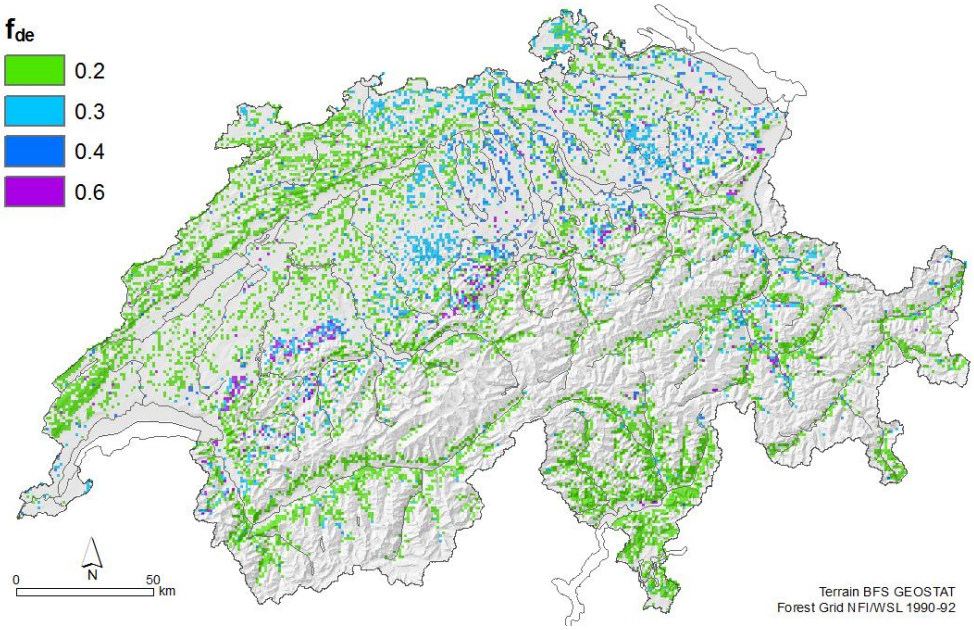
- Soil types: for sandy soils without gleyic features or loess a value of 0.0–0.1, for sandy soils with gleyic features 0.5, for clay soils 0.7, for peat soils 0.8.
- Soil drainage status: excessive 0.0, good 0.1, moderate 0.2, imperfect 0.4, poor 0.7, very poor 0.8.

For calculating $CL_{nut}(N)$ at the 1 x 1 km² raster of the National Forest Inventory), f_{de} was determined according to wetness information from the digital soil map BEK (SFSO 2000) as shown in Table 6 and Figure 14.

Tab. 6 > Values of f_{de} selected for different classes of soil wetness

Wetness class BEK	Description	Depth of water logged horizon	f_{de}
0	Unknown	-	0.2
1	No groundwater	-	0.2
2	Moist	below 90 cm, but capillary rise	0.3
3	Slightly wet	60–90 cm	0.4
4	Wet	30–60 cm	0.6
5	Very wet (not occurring on the digital map)	<30 cm	0.7

Fig. 14 > Denitrification fraction values used for forest sites for the SMB method applied on the 1 x 1 km² raster



3 > Mapping Nitrogen Deposition

3.1 Modelling Approach

Nitrogen **deposition** (N_{dep}) is defined as the sum of reduced (NH_x) and oxidised (NO_y) nitrogen compounds that are deposited by atmospheric processes to a specified receptor of the environment in a specified time period.

Accordingly, nitrogen deposition is used to compute the **exceedance** of critical loads, of empirical critical loads (Ex_{emp}[N]) as well as of critical loads calculated with the SMB method (Ex_{nu}t[N]):

$$Ex_{emp}(N) = N_{dep} - CL_{emp}(N) \quad (3.1)$$

$$Ex_{nut}(N) = N_{dep} - CL_{nut}(N) \quad (3.2)$$

So far, it is not possible to define separate critical loads for the deposition of NH_x-N and NO_y-N. Therefore a total N deposition is used for the calculation of exceedances. The exceedance is time-dependent as it is related to the reference period of the deposition. On the other hand, the critical loads are assumed to be time-invariant.

While the critical load computations (see Chapter 2) are based on methods described in detail in the UNECE mapping manual, the methods for computing depositions were developed specifically for Switzerland. Corresponding proposals in the mapping manual are more on a conceptual level (UNECE 2016, Chapter 2).

In the past, transboundary air pollutant transport and depositions, which are required for modelling abatement scenarios for the UNECE region, have so far been calculated at the European level with 50 x 50 km² resolution (EMEP 2012). Recently, EMEP has carried out a transition to a new grid system with a resolution of 0.1° x 0.1° longitude latitude.

Nitrogen deposition modelling in Switzerland

For national maps, the aim was to estimate ecosystem-specific depositions for the whole area of Switzerland with a high spatial resolution (1 x 1 km² grid or finer), thus, also accounting for the influence of local emission sources. Atmospheric nitrogen deposition was mapped using a pragmatic approach that combines emission inventories, statistical dispersion models, monitoring data, spatial interpolation methods and inferential deposition models. The following nitrogen compounds and deposition paths were considered in the model:

> wet deposition of nitrate (NO_3^-) and ammonium (NH_4^+);
> dry deposition of gaseous ammonia (NH_3), nitrogen dioxide (NO_2) and nitric acid (HNO_3);
> dry deposition of particulate nitrate and ammonium (secondary aerosols).

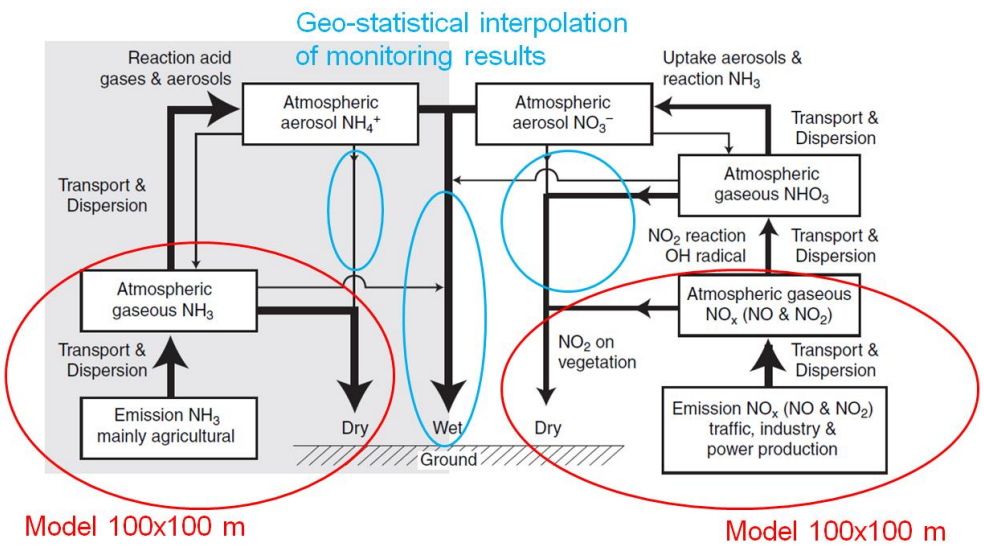
As shown in Figure 15, concentrations of the primary gaseous pollutants (NH_3 and NO_2) are modelled by means of emission maps and statistical dispersion models with a resolution of 100 x 100 m². Thus, the high spatial variability of these pollutants nearby emission sources can be reflected in the models. The concentrations of secondary pollutants (aerosols, HNO_3) as well as the concentrations of N in precipitation are derived from field monitoring data by applying geo-statistical interpolation methods.

Wet deposition is calculated by combining the concentration field of N compounds in rainwater with the precipitation amount (5-year-averages), see Chapter 3.2.

"Inferential" models are used for assessing the dry deposition of gases and aerosols: The concentration maps are multiplied with deposition velocities (V_{dep}), which depend on the reactivity of the pollutant, the surface roughness and climatic parameters (Chapter 3.3). Values of V_{dep} were taken from literature. "Occult" deposition through cloud water or fog is not taken into account.

Maps of emissions, concentrations and deposition were calculated for the years 1990, 2000, 2007 and 2010. For 2000, 2007 and 2010 identical methods were used, i.e. it is a homogenous time series. For the year 1990, a somewhat modified method adapted to the availability of data was applied. The following chapters present the approach and input data used for the year 2010.

Fig. 15 > Deposition modelling scheme



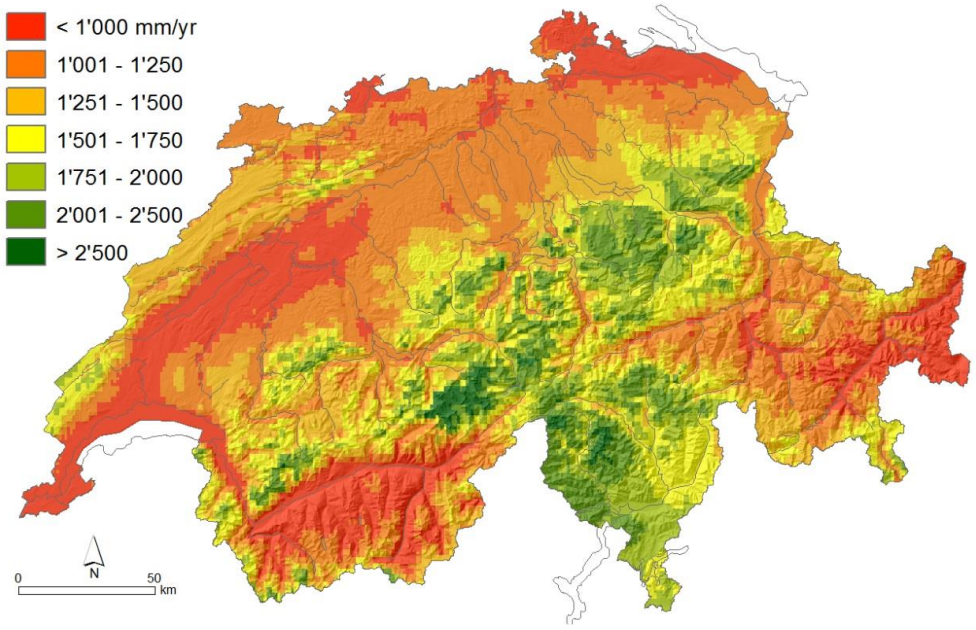
based on figure by Hertel et al. 2011

3.2 Wet Deposition

Wet deposition was obtained by multiplying the annual precipitation rate with mean concentrations of soluble inorganic N compounds in precipitation. For precipitation and concentrations a 5-year-average (2008–2012) was used. Maps with interpolated precipitation amounts were supplied by MeteoSwiss (2013, see Figure 16). They are based on nearly 300 monitoring stations.

In regions with high precipitation, the calculated deposition can be overestimated as the scavenging effect (wash-out of gases and particles by rain) gets smaller. Therefore, precipitation was limited to maximum of 1800 mm a^{-1} in the deposition computation.

Fig. 16 > Precipitation, average 2008–2012, 1 x 1 km² raster



MeteoSwiss 2013

Concentration of nitrogen compounds in rainwater

For mapping N concentrations in rainwater, different approaches are used in Northern and Southern Switzerland. In Northern Switzerland, data of the National Air Pollution Monitoring Network (BAFU 2015) on wet deposition imply that there is no clear geographic pattern in rainwater concentrations of N at low altitudes, but an obvious decrease of concentrations with increasing altitude. However, monitoring stations with wet-only measurements are relatively scarce in Northern Switzerland and especially at altitudes above 1200 m there is only little information available at the moment (some more stations were recently put into operation, see Seitler et al. 2016). Therefore, we roughly assumed a constant concentration of 0.70 mg N l^{-1} (0.42 mg NH_4-N + 0.28 mg NO_3-N l^{-1}) below 800 m altitude, a linear decrease of 70% between 800 and 3000 m altitude and again a constant concentration above 3000 m. The base concentration below 800 m is the 5-year-average of the stations Payerne and Dübendorf situated on the Central Plateau at approximately 450 m altitude.

The assumptions related to the decrease in concentrations between 800 and 3000 m altitude were adjusted during the last years based on comparisons with measurements carried out at higher altitudes on Swiss Long-term Forest Ecosystem Research plots (Thimonier et al. 2005) and on Alpine grassland (Hiltbrunner et al. 2005).

In Southern Switzerland, there are several stations run by the Canton Ticino besides NABEL and neighbouring Italian stations. Thus, the wet-only monitoring network is relatively dense. Concentrations in rainwater and deposition maps were calculated by regression models considering longitude, latitude and altitude. They were initially developed by Barbieri & Pozzi (2001) and updated by Steingruber & Colombo (2010) and Steingruber (2014). For the period 2008–2012, ammonium and nitrate concentrations in precipitation can be calculated for the region of the Canton Ticino as follows:

$$C_{wet}(NH_4^+) = -22.4 + 0.0001238 \cdot x - 0.0002581 \cdot y - 0.00461 \cdot z \quad (3.3)$$

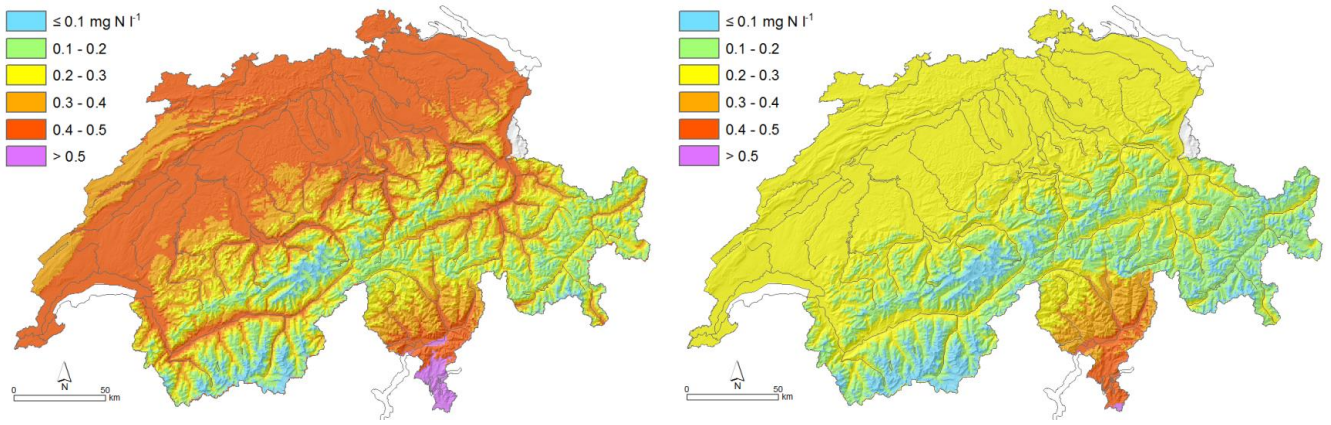
$$C_{wet}(NO_3^-) = -66.4 + 0.0001581 \cdot x - 0.0001366 \cdot y - 0.00292 \cdot z \quad (3.4)$$

where:

$C_{wet}(N)$	*concentration of N-compound in rainwater [meq m^{-3}].*
x, y	*longitude/latitude in Swiss projection LV03*[11] *(m).*
z	*altitude (m).*

The resulting concentration maps are shown in Figure 17.

Fig. 17 > Concentration in rainwater of ammonium (left) and nitrate (right), average 2008–2012, 1 x 1 km² raster



[11] www.swisstopo.admin.ch/internet/swisstopo/en/home/topics/survey/sys/refsys/projections.html

3.3 Dry Deposition of Gases

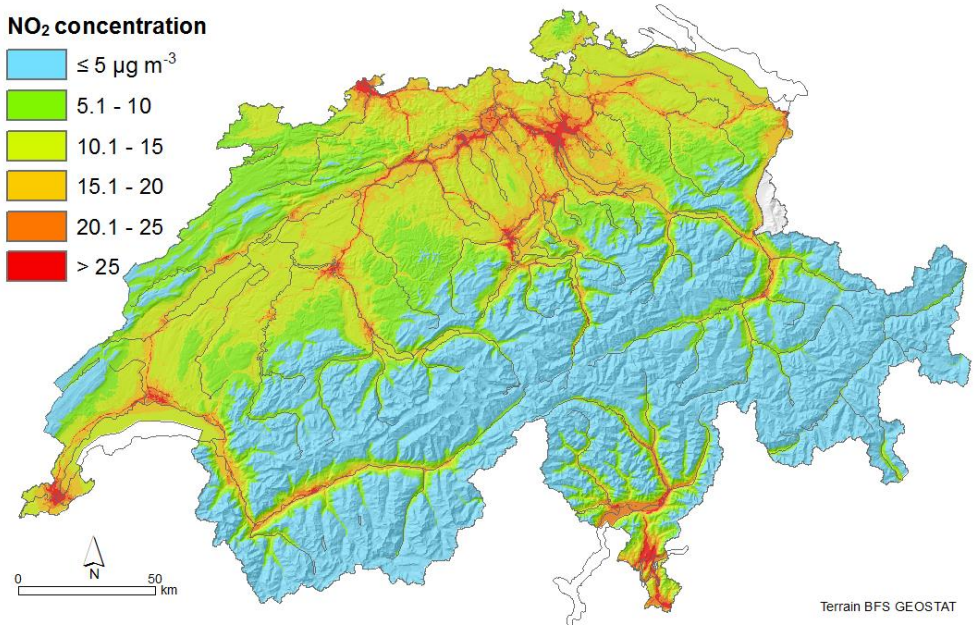
Dry deposition of NH_3, NO_2 and HNO_3 gases was calculated on the basis of modelled air concentrations (annual means) and average deposition velocities (V_{dep}) applying small-scale "inferential models" (UNECE 2016, Chapter 2). Dry deposition is inferred by multiplying the concentration with the deposition velocity of the component of interest. V_{dep} can be calculated using a resistance model in which the vertical transport to the ground and the absorption of the component by the surface is described. For calculating the Swiss deposition maps, average values of V_{dep} from literature were used representing typical conditions for different pollutants and land-use types.

For NH_3 and NO_2 the air concentrations were calculated applying an emission-dispersion approach (see Fig. 15). Emissions, dispersion and concentrations of NH_3 were mapped on a grid of 100 x 100 m². The NH_3 model is presented in detail in Chapter 3.5.

Nitrogen dioxide concentration map

NO_2 concentrations for the year 2010 were available on a 100 x 100 m² raster from the Federal Office for the Environment (FOEN 2011), see Figure 18. They were calculated on the basis of a comprehensive emission inventory for transport, industry, households/commerce and agriculture/forestry.

Fig. 18 > NO_2 concentration, annual mean 2010, 100 x 100 m² raster



Measurements of HNO_3 are very rare. Therefore, the concentration map for HNO_3 was derived from existing maps of air humidity, temperature, ozone concentrations and NO_2 concentrations applying an empirical relationship developed by ICP Materials (UNECE 2005):

3.5

$$HNO_3 = 516\ e^{-3400\ /\ (T+273)}\ (Rh\ [O_3]\ [NO_2])^{0.5}$$

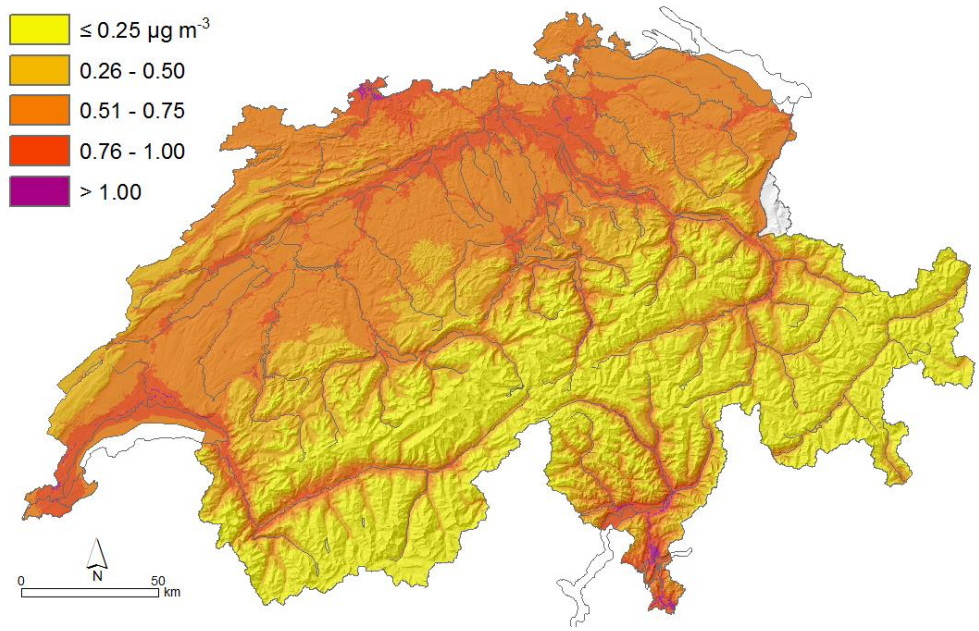
Where:

T	*temperature in° C, annual mean.*
Rh	*relative humidity in percent, annual mean.*
[O_3]	*ozone concentration in µg m^{-3}, annual mean.*
[NO_2]	*nitrogen dioxide concentration in µg m^{-3}, annual mean.*

Nitric acid concentration map

The input maps for temperature, humidity and ozone were produced for the modelling of corrosion rates (Reiss et al. 2004) with a resolution of 250 x 250 m² using geo-statistical interpolation methods. The resulting annual mean concentrations of HNO_3 are between 0 and 1.2 µg m^{-3} (Figure 19) which is in a range confirmed by existing monitoring results in Switzerland (Thöni & Seitler 2010).

Fig. 19 > HNO_3 concentrations, 250 x 250 m² raster



Annual mean 2000

Deposition velocities (V_{dep}) can be calculated using a resistance model in which the vertical transport to the ground and the absorption of the component by the surface is described. For calculating the Swiss deposition maps, average values of V_{dep} from literature were used representing typical conditions for different pollutants and land-use types. Hertz & Bucher (1990) compiled various V_{dep} for ecosystems in Switzerland based on literature. In the mapping study by FOEFL (1996) those values were used. Later they were adjusted or differentiated based on more recent studies.

Deposition velocities of ammonia

For NH_3, typical values of V_{dep} are obtained from Sommer and Jensen (1991), Sutton et al. (1992), Sutton et al. (1994), Fangmeier et al. (1994), Asman (2002), Krupa (2003), Loubet et al. (2009) and Cape et al. (2009). Values given by Cape et al. (2009) for conditions in the United Kingdom are in a range of 16–32 mm s^{-1} for low vegetation and 33–48 mm s^{-1} for high vegetation. For Switzerland, somewhat lower values are used (Table 7):

- 22–30 mm s^{-1} for forests; coniferous forests have generally a rougher surface and no needle loss in winter which implicates higher annual V_{dep} in comparison with deciduous forests.
- 10–20 mm s^{-1} for low vegetation; ammonia is absorbed faster by wet surfaces than under dry conditions. Therefore, unproductive vegetation mainly consisting of wetlands and shrubs has a higher V_{dep} than dry grassland. On fertilised areas (cropland and grassland) V_{dep} can be interpreted as an annual net deposition velocity. During and several days after the application of nitrogen containing fertilizers these areas are an emission source, while they act as a sink during the rest of the year.
- For areas with no or little vegetation, V_{dep} between 5 and 8 mm s^{-1} are used.

Deposition velocities of nitrogen dioxide

V_{dep} for NO_2 are lower than for NH_3. On the basis of values indicated by Hertz & Bucher (1990), the Harwell Laboratory (1990), Lövblad & Erisman (1992), Hornung et al. (1994), Duyzer & Fowler (1994), Hesterberg et al. (1996) and Wu et al. (2011), values between 1.5 and 4.0 mm s^{-1} are applied (Table 7).

Deposition velocity of nitric acid

For HNO_3 the deposition velocity is set to 15 mm s^{-1} independent of land use type. This is a conservative estimate. This V_{dep} is based on values listed by Harwell Laboratory (1990), Riedmann & Hertz (1991), Lövblad & Erisman (1992), Andersen & Hovmand (1995) and Wesley & Hicks (2000). According to Duyzer & Fowler (1994) the canopy resistance to uptake of HNO_3 is negligible, thus leading to a very efficient deposition to the cuticle and to a dominant stomatal uptake.

Tab. 7 > Deposition velocities of gaseous pollutant compounds

Pollutant	Land-use type	Vdep mm s^{-1}	Remarks
NH_3	coniferous forest	30	>90% coniferous trees
	mixed forest	26	11–90% coniferous trees
	deciduous forest	22	>90% deciduous trees
	unproductive vegetation	20	includes wetlands (fens, bogs)
	meadows, pastures	12	includes dry grassland (TWW)
	cropland, grassland	10	intensely managed, fertilized
	settlements	8	buildings/surroundings, transportation areas
	surface water	8	lakes, rivers
	bare land	5	rocks, sand, glaciers
NO_2	coniferous forest	4	>50% coniferous trees
	deciduous forest	3	>50% deciduous trees
	non-forest	1.5	
HNO_3	all	15	

The land-use type is defined on the basis of the National Forest Inventory (WSL 1990/92, 1 x 1 km² grid), the Swiss Land-use Statistics (SFSO 2005, ha-grid) and a ha-grid indicating the forest type (mixing ratio of coniferous and deciduous trees, SFSO 2004).

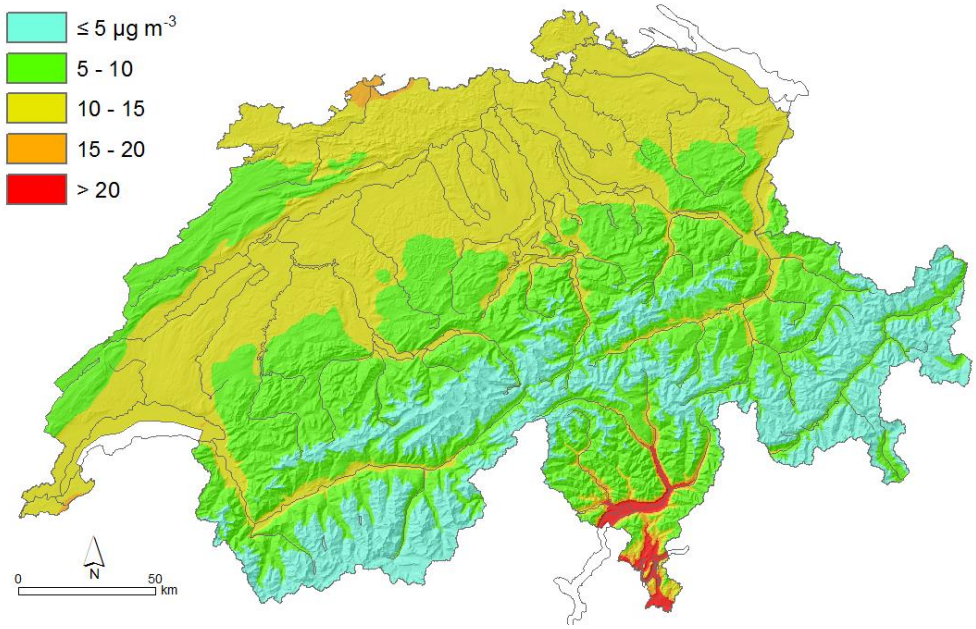
3.4 Dry Deposition of Aerosols

Dry deposition of aerosols includes (1) the gravitational sedimentation of aerosols not bound to precipitation, and (2) the "interception", which is the filtering effect of the vegetation through horizontal impact of aerosols. Sedimentation and interception are treated together since they both depend on wind speed and surface properties of the vegetation layer. The dry deposition is calculated by multiplying the concentration in the air with an altitude and receptor dependent deposition velocity.

Concentrations of nitrate and ammonium in particulate matter

The concentration of particulate nitrate and ammonium was derived from modelled and mapped concentrations of particulate matter PM10 (SAEFL 2003). The spatial pattern was determined by selecting only the model layers containing secondary aerosols (Figure 20) assuming that primary particles hardly contain any nitrogen. According to chemical analysis of PM10 at sites on the Central Plateau, the mean concentration was approximately 2.0 µg NH_4^+ m^{-3} and 2.8 µg NO_3^- m^{-3} (SAEFL 2003). These concentrations add up to approximately 37% of the modelled average secondary aerosol concentrations in the Central Plateau (13 µg m^{-3}). It was assumed that this portion of 37% is constant in all regions of the country.

Fig. 20 > Concentrations of secondary PM10, 200 x 200 m² raster



after SAEFL 2003

Deposition velocities of aerosols

V_{dep} are average values obtained from literature (e.g. Lövblad & Erisman 1992, Asman & van Jaarsveld 1992, Lövblad et al. 1993; Duyzer (1994), Fangmeier et al. 1994, Hesterberg et al. 1996, Loubet et al. 2009). An estimate of the altitude dependence of V_{dep} for Switzerland was observed by Riedmann & Hertz (1991) as follows (see Table 8):

3.6

$$V_{dep}(900m) = 2 \bullet V_{dep}(450m)$$

The increase of V_{dep} with altitude can be explained by the fact that the average wind speed also increases strongly, rising from 450 m to 900 m a.s.l in the Central Plateau. Thereby the filtering effect of vegetation is reinforced. Between 900 m and 2000 m an increase also exists, although it is smaller.

The additional filtering effect of forests is taken into account by using deposition velocities that are 1.5–2.5 times higher than for open fields (see Table 8).

Tab. 8 > Deposition velocities of aerosols containing nitrate and ammonium depending on altitude and land use

Altitude	Land-use type	V_{dep} mm s^{-1}
≤ 400 m	coniferous forest	2.5
	mixed forest	2.0
	deciduous forest	1.5
	non-forest	1.0
≥ 800 m	coniferous forest	5.0
	mixed forest	4.0
	deciduous forest	3.0
	non-forest	2.0
400–800 m	linear interpolation	

3.5 Mapping Ammonia

3.5.1 Emissions of Ammonia

Agriculture is the main source of NH_3 accounting for about 92% of total Swiss ammonia emissions, other anthropogenic sources account for 7% and natural sources for 1% (Table 9).

Tab. 9 > Ammonia emissions for different source categories in 2010

Source category	Emission (kt NH_3-N/a)	Percent
Housing/hardstandings	15.0	28.4%
Storage of manure	7.3	13.8%
Application of manure	20.1	38.0%
Grazing	1.2	2.2%
Total livestock	**43.5**	**82.4%**
Mineral fertilizers	2.0	3.8%
Organic recycling fertilizers	0.4	0.8%
Cropland, grassland, alpine pastures	2.4	4.5%
Total plant production	**4.8**	**9.1%**
Total agriculture	**48.3**	**91.5%**
Commerce and industry	0.5	0.9%
Transport	2.3	4.4%
Households	0.8	1.5%
Waste treatment	0.3	0.6%
Total non-agriculture	**3.9**	**7.4%**
Natural sources	0.6	1.1%
Total emission	**52.8**	**100.0%**

after Kupper et al. 2013

Mapping agricultural ammonia emissions

The emission maps were calculated with a resolution of 100 x 100 m². The emissions from animal husbandry were modelled with livestock statistics of the year 2010 using a 'bottom-up' approach (i.e. with georeferenced activity data and emission factors). The other emission categories (crop farming, industries, traffic, households, waste treatment and natural sources) were spatially allocated in a top-down approach to the relevant categories of a land-use map. The procedure is summarised in the following sections.

(1) The SFSO (2013) supplied the livestock per farm as well the locations of 61 000 farms (coordinates rounded to hectometres). Hence the emissions from animal husbandry (43.5 kt NH_3-N) were calculated for each farm (f) as:

3.7

$$E(f)_s = \sum_a (N(f)_a \cdot EF_{acs})$$

where:

$E(f)_s$	*NH_3-emissions on the farm f at the emission stage s [kg NH_3-N a^{-1}].*
s	*emission stage (housing, manure storage, manure application, grazing)*
$N(f)_a$	*number of animals in category a on the farm f.*
a	*animal category (see Table 10).*
EF_{acs}	*emission factor of animal category a in farm class c at the emission stage s [kg NH_3-N a^{-1} $animal^{-1}$].*
c	*farm class; combines geographic region, altitude zone and production type.*

For each farm, the livestock numbers were multiplied with emission factors that are stratified into 24 animal categories, 6 emission stages and 32 farm classes. The fully stratified emission factors (not shown) were calculated by Bonjour (2013). Table 10 presents an overview of the total animal numbers in Switzerland, the mean emission factors (emission per animal) for the 6 emission stages (housing/hardstandings, storage of liquid and solid manure, application of liquid and solid manure, grazing) and the resulting total emissions from animal husbandry. I.e. the emission factors of the 32 farm classes were averaged in Table 10.

Farm class accounts for the differences in production techniques in various regions of the country. It is derived by combining three geographic regions (Central, Eastern and Western/Southern Switzerland), three altitude zones (valley, hill and mountain) and five production types (arable farms, cattle farms, pig/poultry farms, mixed farms and other farms) as shown by Kupper et al. (2013).

Tab. 10 > Emission factors, number of animals and total NH_3-emissions for 24 animal categories in 2010

	Emissionsfactor (kg NH_3-N / a per animal)								
Animal category	Housing, hard standings	Storage liquid manure	Storage solid manure	Application liquid manure	Application solid manure	Grazing	**Total**	Number of animals	Emision (kt NH_3-N/a)
Dairy cows	9.50	4.54	1.83	16.40	2.85	0.88	**36.00**	589 024	21.20
Suckling cows	8.87	2.60	1.50	8.15	2.59	1.29	**25.00**	111 291	2.78
Dairy followers <1 yr	2.69	0.62	0.95	2.53	1.40	0.27	**8.45**	226 352	1.91
Dairy followers 1–2 yr	3.63	1.17	0.95	3.89	1.46	0.68	**11.80**	212 778	2.51
Dairy followers >2 yr	4.76	1.50	1.36	6.44	2.20	0.84	**17.10**	119 163	2.04
Pre beef-fattening calves	3.96	0.63	1.27	4.01	1.69	0.35	**11.90**	88 095	1.05
Beef cattle	4.21	1.29	1.42	3.76	1.84	0.07	**12.60**	145 084	1.83
Beef calves	1.54	0.14	1.30	0.56	1.73	0.00	**5.27**	99 446	0.52
Farrowing sows	10.00	2.16	0.01	4.99	0.00	-	**17.20**	33 508	0.58
Dry sows	6.34	1.02	0.00	2.18	0.00	0.00	**9.54**	106 070	1.01
Piglets <25 kg	1.11	0.21	0.02	0.54	0.00	0.00	**1.89**	349 206	0.66
Boars	5.56	0.89	0.04	2.07	0.00	0.02	**8.58**	3 685	0.03
Fattening pigs	3.17	0.48	0.00	1.67	0.00	0.00	**5.33**	788 149	4.20
Laying hens	0.14	-	0.04	-	0.08	0.02	**0.28**	2 438 051	0.68
Poultry growers	0.05	-	0.02	-	0.03	0.00	**0.10**	925 522	0.09
Broilers	0.06	-	0.01	-	0.06	0.00	**0.12**	5 580 103	0.67
Turkeys	0.18	-	0.05	-	0.14	0.01	**0.38**	58 074	0.02
Other poultry	0.08	-	0.04	-	0.08	0.00	**0.21**	23 153	0.00
Horses >3 yr	4.33	-	2.32	-	2.59	0.54	**9.78**	53 441	0.52
Horses <3 yr	3.70	-	1.98	-	1.95	0.71	**8.33**	8 672	0.07
Ponies, donkeys, mules	1.56	-	0.73	-	1.19	0.16	**3.65**	20 407	0.07
Sheep	1.19	-	0.75	-	0.85	0.26	**3.05**	228 178	0.70
Milk sheep	2.13	-	1.00	-	1.38	0.24	**4.76**	12 362	0.06
Goats	1.68	-	1.11	-	1.01	0.08	**3.88**	54 739	0.21
Total									**43.5**

after Kupper et al. 2013

(2) It was assumed that the emissions from housing, hardstandings and manure storage are located in the same hectare-cell as the farm building (total 22.3 kt NH_3-N a^{-1}, see Tab. 9).

(3) The emissions from grazing and manure application (total 21.3 kt NH_3-N a^{-1}, see Tab. 9) were distributed over the agricultural areas within the municipality.

Municipality boundaries are used here as a proxy for the land managed by the farms as it is not known, on a national level, which parcels belong to a specific farm. The municipality areas are available in digital form at swisstopo (2010). The size of the ~2800

municipalities varies from about 3 km² in the Plateau to more than 100 km² in alpine regions (average 14 km²). Inside each municipality, the land use-map (SFSO 2005) with a resolution of one hectare is used to enhance the plausibility of the spatial emission pattern. This is done by weighting the land-use categories as follows:

- Weight = 1: arable land, meadows, farm pastures, vineyards and orchards.
- Weight = 0.25: open forest (on agricultural areas), brush meadows/farm pastures, mountain meadows and favourable alpine pastures. These categories are assumed to emit much less ammonia than arable land and regular meadows because they are often situated in relatively remote areas or because the season for grazing is relatively short.
- Weight = 0: all other land-use categories are excluded from the area sources. Furthermore, all areas protected by the Federal Inventories of Raised and Transitional Bogs and of Fenland (see Chapter 2.2.3) are excluded as fertilizing is generally not allowed.

(4) The portion of emissions produced by animals which spend the summer (90 days) up on the alps amounts to 1.7 kt NH_3-N. On the one hand, this emission quantity was subtracted from the emissions of the respective farms (with municipalities as spatial reference). On the other hand, these emissions were redistributed over the land-use type “alpine pastures” for the whole of Switzerland. The portion of summered animals was calculated from livestock statistics 2000 (SFSO 2013).

(5) Emissions from the application of mineral fertilizer and organic recycling fertilizer (total 2.4 kt NH_3-N a^{-1}, see Tab. 9) were distributed over the land-use categories arable land, meadows, farm pastures, vineyards and orchards.

(6) Emissions on agricultural land arising from processes in the plant layer (total 2.4 kt NH_3-N a^{-1}, see Tab. 9) were allocated according to Kupper et al. (2013):

- An emission factor of 2.0 kg NH_3-N ha^{-1} was applied on land-use categories arable land, meadows, farm pastures, vineyards and orchards. The total area is 1 060 kha and the resulting emission is 2.1 kt NH_3-N a^{-1}.
- An emission factor of 0.5 kg NH_3-N ha^{-1} was applied on land-use categories mountain meadows, favourable alpine pastures, remote/steep alpine meadows and remote/steep alpine pastures. The total area is 538 kha and the resulting emission is 0.3 kt NH_3-N a^{-1}.

Ammonia emissions from industry, commerce, traffic, households and waste treatment

(7) Emissions from commerce and industry (0.5 kt NH_3-N a^{-1}) were distributed over the land-use categories industrial buildings and industrial grounds.

(8) The ammonia emissions from transport were mapped by a bottom-up approach for the year 2000. It was based on a traffic model including the main road-networks and zonal traffic (SAEFL 2004). This emission map was proportionally rescaled to the total transport emission in 2010 (2.3 kt NH_3-N a^{-1}).

(9) Emissions from households (0.8 kt NH_3-N a^{-1}) were distributed over the land-use categories residential areas, agricultural buildings and garden allotments.

(10) Emissions from treatment of waste-water and solid waste (0.3 kt NH_3-N a^{-1}) were distributed over the land-use categories ‘waste water treatment plants’ and dumps.

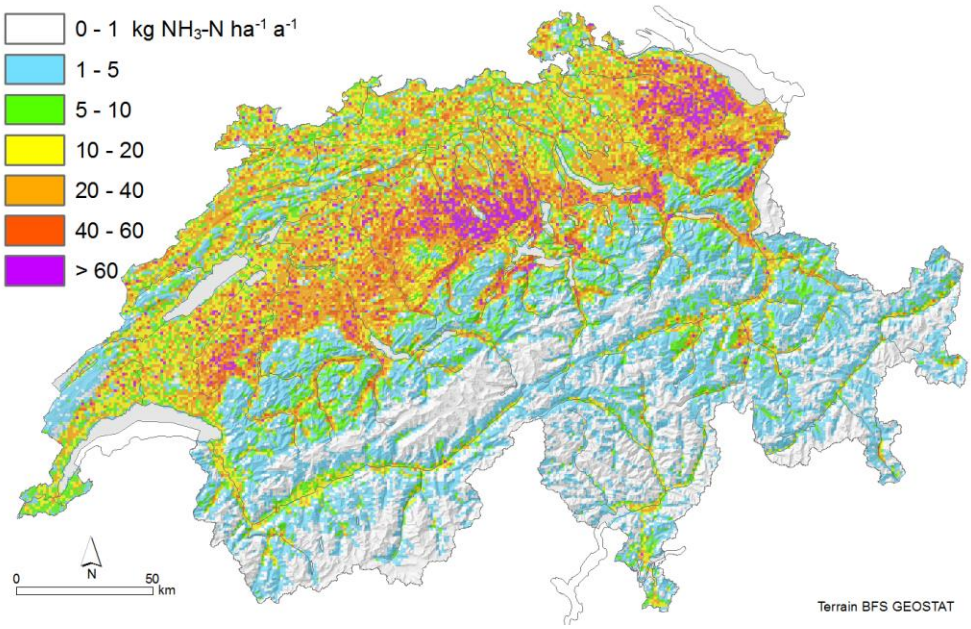
Natural ammonia emissions

(11) Natural emissions (0.6 kt NH_3-N a^{-1}) originating from soil processes on natural grassland as well as from wild animals were distributed over the land-use categories forests, woods and unproductive grass/shrubs.

The resulting emission map is shown in Figure 24.

Fig. 21 > Ammonia emissions 2010, modelled on 100 x 100 m² raster

Resolution displayed on map: 1 x 1 km².



3.5.2 Ammonia Concentration

Mapping ammonia concentrations

A statistical dispersion model with a resolution of 100 x 100 m² was applied to calculate annual mean concentrations of ammonia in the air. It includes all emission sources at a maximum distance of 50 km. For each raster-cell the concentration C (µg m^{-3}) induced by the emissions from surrounding sources was calculated as:

3.8

$$C = \sum_i (17/14 \cdot E_i \cdot p(D_i))$$

where:

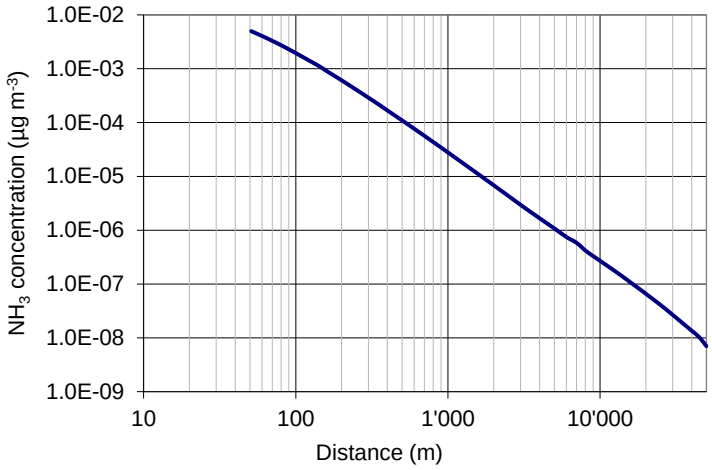
i	*denotes all sources (raster-cells) within a radius of 50 km.*
E_i	*NH_3-emission from raster-cell i (kg NH_3-N a^{-1}), see Chapter 3.5.2.*
17/14	*conversion from [kg NH_3-N] to [kg NH_3].*
D_i	*Distance to source i (m).*
$p(D_i)$	*average dispersion-profile; concentration as a function of D_i.*

p(D) is a normalised average distance-function providing the annual mean concentration (µg m^{-3}) induced by one kg NH_3 a^{-1} emitted from a source at a distance of D (see Figure 22 and Annex). The function was calculated by Asman & Jaarsveld (1990) using an atmospheric transport model. The model was applied and tested in several countries (The Netherlands, United Kingdom, Belgium, Denmark and Sweden) and in Europe (Asman & Jaarsveld 1992). In Switzerland, the integrated distance-function shown in Figure 22 and Equation 3.8 was used the first time by Rihm & Kurz (2001) to calculate NH_3-concentration maps; the comparison with measured NH_3-concentrations was promisingly good. Thus, the method was further developed and used for mapping NH_3 in Switzerland by Thöni et al. (2005), Rihm et al. (2009) and EKL (2014).

The function p(D) integrates effects of several atmospheric processes such as dilution by turbulence, deposition on the ground and chemical transformation of ammonia to ammonium. It is related to a point source at a height of 3 m above ground and the resulting concentrations are given for a reference height of 1 m above ground. In the present study, it was assumed that the sources and the receptor points are situated in the centre of the 100 x 100 m² cells. For calculating the concentration in a raster-cell induced by the emission of the same cell (where D = 0 m), a distance of 50 m was used in p(D).

Fig. 22 > NH_3 concentration as a function of the distance to a source emitting 1 kg NH_3 a^{-1} for distances between 50 m and 50 km

See Annex.



by Asman & Jaarsveld 1990

In this study, the calculation was made with a resolution of 100 x 100 m² for distances below 4 km. Due to computing performance, distances between 4 km and 50 km were calculated on the basis of spatially aggregated emission-data with a resolution of 1 x 1 km².

Furthermore, two different climatic regions were defined for the calculation of distances below 4 km:

> In the first region (covering the Alps and Southern Switzerland), the function p(D) was applied rotation-symmetrically (Figure 23, left).
> In the second region (covering the Jura Mountains and the Swiss Plateau) the p(D) function was modified according to prevailing wind directions in this area (Figure 23, right); i.e. the concentration is calculated as a function of distance and azimuth between source and receptor. The modification was based on the region-specific dispersion profiles developed for NO_2 (FOEN 2011).

The resulting map of ammonia concentrations in 2010 is shown in Figure 24.

Fig. 23 > Schematic dispersion profiles for distances 50–4000 m

Rotation-symmetric use of p(D) in the Alps and Southern Switzerland.	*Modified p(D) function according to prevailing wind directions in the Jura/Plateau region.*

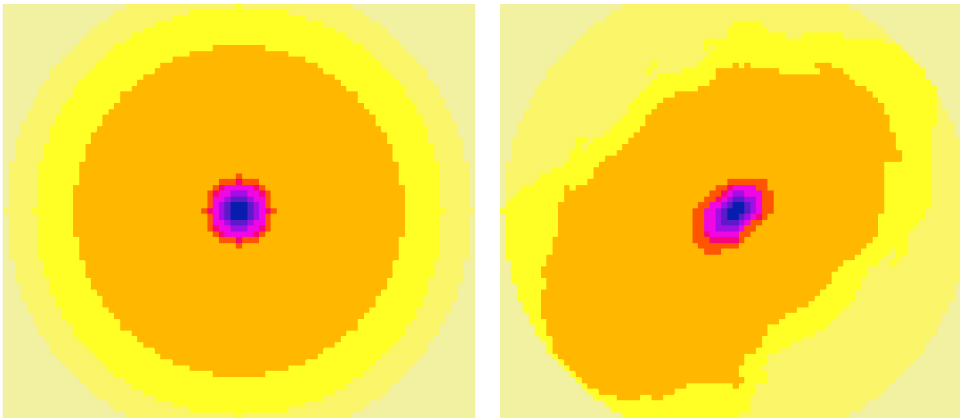
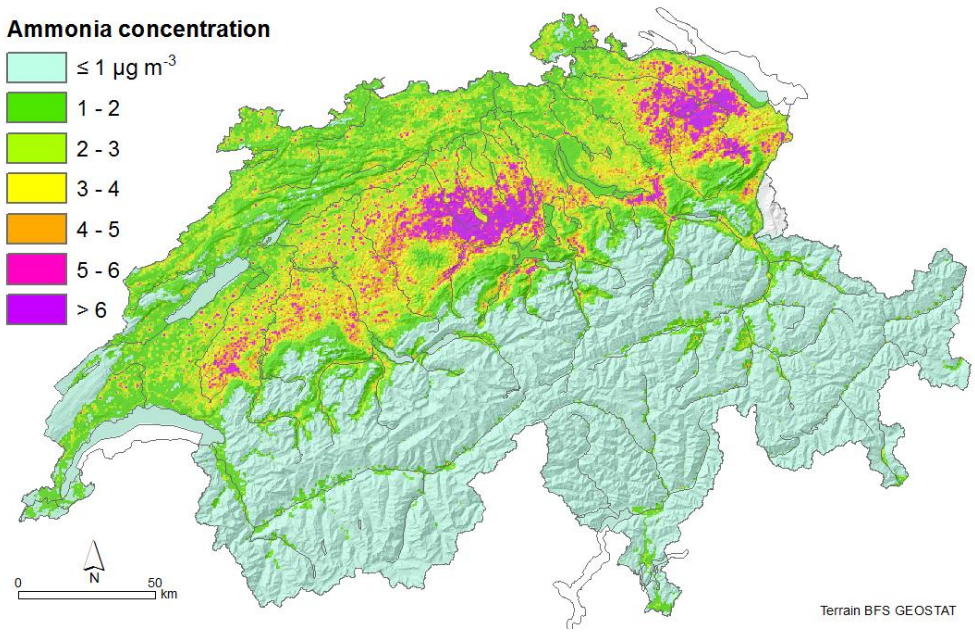


Fig. 24 > Map of NH_3 concentration in 2010, 100 x 100 m² raster



3.5.3 Validation of the Ammonia Maps

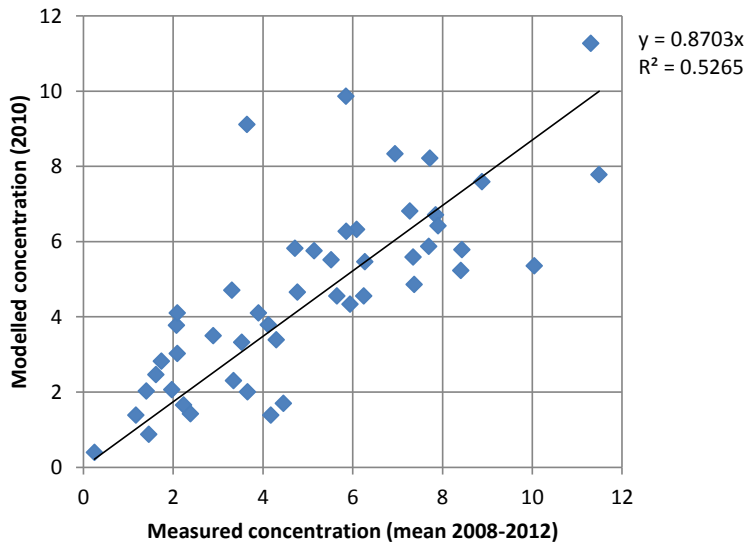
Comparison of modelled and measured ammonia concentrations

The modelled concentrations can be validated by comparing with measurements provided by Seitler & Thöni (2016). These measurements show that there is an inter-annual variation of concentrations due to climate (higher concentrations in warm years) but there is no significant trend between 2000 and 2014. For the comparison shown in Figure 25, 48 monitoring sites with complete time-series 2008–2012 were selected and the average of the 5-year period was used as the modelled values do not include climatic variations.

The trend-line in Figure 25 shows that on the average modelled concentrations are somewhat below the measured values. In a detailed statistical analysis, Locher (2014) revealed that mainly in Alpine valleys and in Southern Switzerland the model values are too low. That study could be a good starting point for further improvements of the model since it highlights possible and promising approaches leading to a somewhat narrower relation between measured and modelled concentrations.

Fig. 25 > Comparison of measured and modelled ammonia concentrations (µg m^{-3})

N=48.



The normalised RMS error value of the data in Figure 25 is 43%. A Monte-Carlo simulation carried out with *a priori* uncertainties for the model parameters identified the distance D and the shape of the dispersion function p(D) as the most critical parameters. Mainly at sites close to point sources (farm buildings) the rasterization to hectare-cells implicates considerable uncertainty of D and is responsible for a part of the scatter visible in Figure 25. In addition, the real position of the sources (housing/ hardstandings, manure storage) is not necessarily identical with the coordinates of the main farm building supplied by SFSO (2013). Of course, such offsets also hold for non-agricultural sources mapped with the top-down approach (see Chapter 3.5.1). Currently, these inherent shortcomings of the emission map and the dispersion model cannot be improved at the national level.

Although the Swiss climate differs from the climate of those regions where Asman & Jaarsveld (1990) calculated the dispersion function p(D) (Fig. 22), the results of the present model correspond quite well to monitoring data (Figure 25). Probably, there were some contrarian effects compensating each other: On one hand, the lower wind speed in Switzerland would lead to higher concentrations on the ground. On the other hand, the well mixed land-use pattern (cropland, grassland, forests, and settlements) and the complex topography lead to a generally high surface roughness on the landscape level and would thus lead to lower concentrations on the ground. Furthermore,

the mixed pattern of sources and sinks in large parts of the country might lead to a relatively high local deposition. These presumptions were tested with the Lagrangian dispersion model AUSTAL[12] on a flat terrain for distances 100–1000 m. With a typical meteorological data set for the Swiss Plateau and appropriate parameterisations of deposition velocity (20 mm s^{-1}) and surface roughness (0.5 m) AUSTAL could reproduce the dispersion function p(D) consistently. Of course, local climatic effects such as thermal and cold air flow are not included in the national model application and remain an important source of uncertainty.

3.6 Mapping Depositions 1990–2010

Time series of nitrogen depositions

Besides the deposition map for the year 2010, also maps for 1990, 2000 and 2007 were produced. The maps 2000–2010 represent consistent time-series. For 1990 however, the mapping method for ammonia emissions was different. In this study, the maps for 1990, 2000 and 2010 are used to calculate exceedances of the critical loads as shown in Chapter 4.3 (deposition map 2007 is not used). The methods for calculating depositions in 1990 and 2000 were very similar to those used for 2010. The most important modifications were:

- Ammonia emissions in 1990 from livestock were mapped by a top-down approach: The total amounts per emission stage given by Kupper et al. (2013) were distributed to the farms proportionally to the livestock units (GVE) of each farm. The location and the livestock units of the farms were supplied by SFSO (2013) for the year 1996 – the first national livestock statistics featuring geographic coordinates.
- Ammonia emissions in 2000 from livestock were mapped on the basis of the livestock statistics 2000 (location of farms and number of animals, SFSO 2013) and with the emission factors developed by Kupper et al. (2013) for the year 2002. For mapping purposes, the livestock statistics 2000 was preferred because in the statistics 2002 the geographic coordinates were incomplete.
- For wet deposition, the 5-year averages of concentrations in rain and precipitation amounts were used, 1988–1992 and 1998–2002, respectively.

On the average, HNO_3 and aerosols contribute less than 10% to the total deposition. For simplification purposes and because of lack of data, these parameters were left constant for the whole period.

[12] http://austal2000.de

4 > Results

4.1 Critical Loads of Nitrogen

The spatial distributions of critical loads are displayed separately for the empirical method and for the SMB method on the maps in Figure 26 and Figure 27, respectively.

The map in Figure 26 is a combination of all selected ecosystems as described in chapter 2.2.3. Only (semi-)natural ecosystems with high conservation importance were included. For each cell of the 1 x 1 km² raster, the critical load value of the most sensitive ecosystem existing in the cell was selected. The map in Figure 28 combines the results of the empirical method and SMB method showing the minimum critical load of both.

Figure 29 presents the critical loads as cumulative frequency distributions.

Fig. 26 > Empirical critical loads of nutrient nitrogen for (semi-)natural ecosystems, $CL_{emp}(N)$

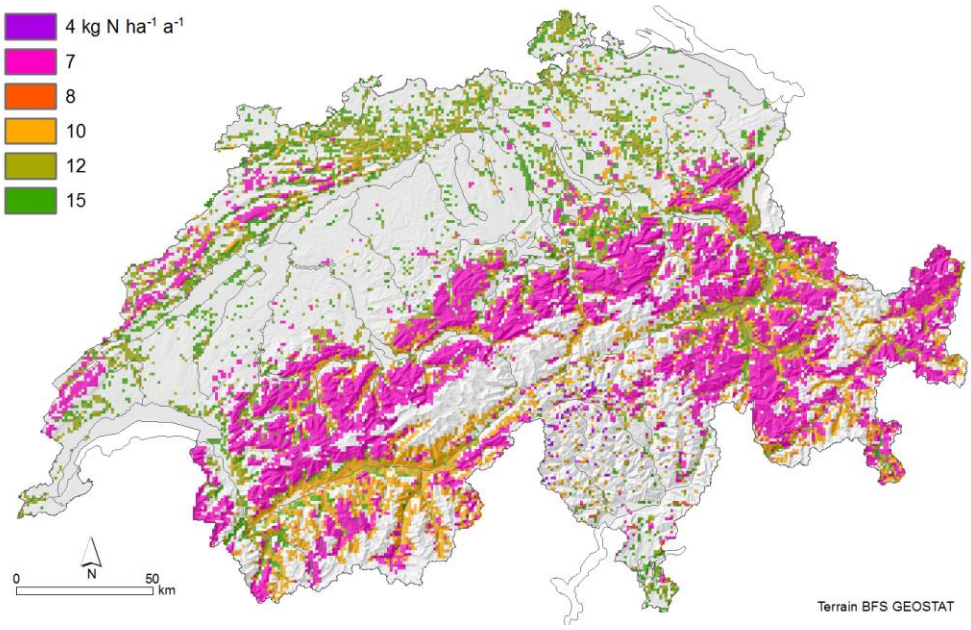


Fig. 27 > Critical loads of nutrient nitrogen for forests, $CL_{nut}(N)$

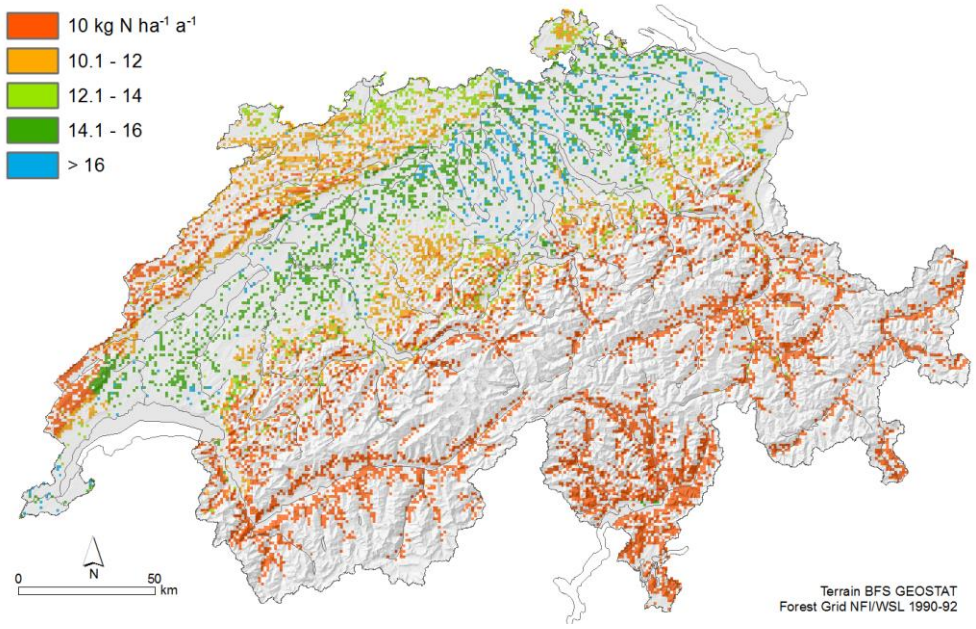


Fig. 28 > Combined critical loads of nutrient nitrogen, minimum of $CL_{nut}(N)$ and $CL_{emp}(N)$ per km²

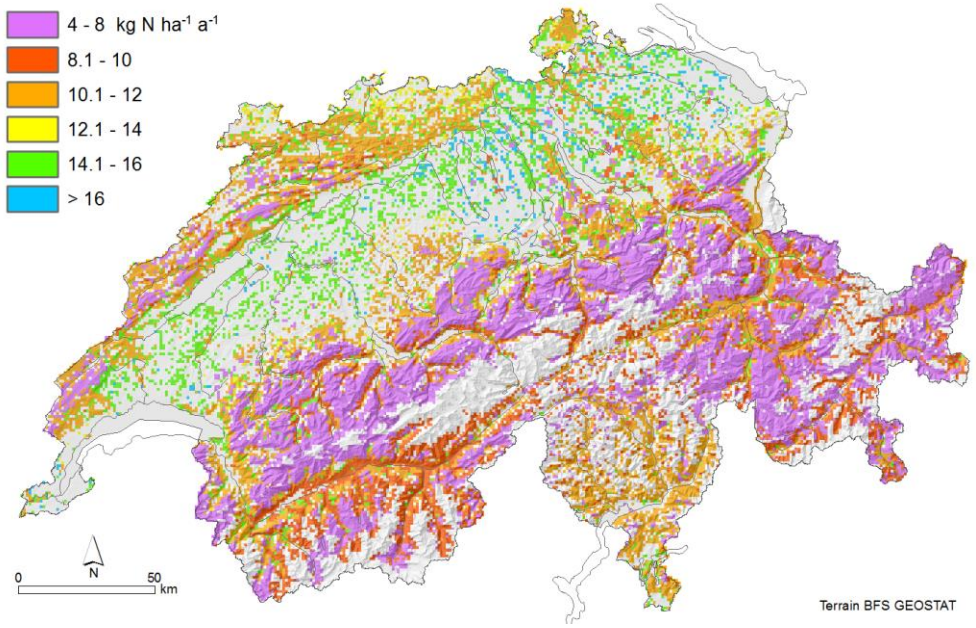
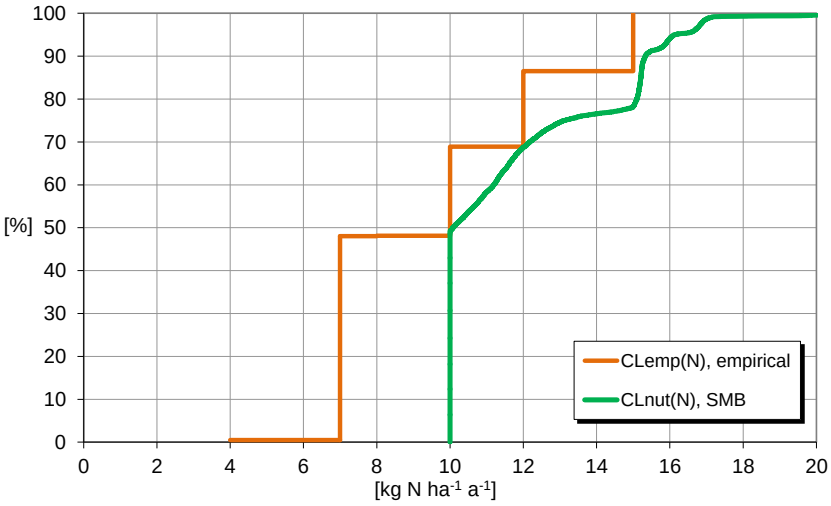


Fig. 29 > Cumulative frequency distributions of critical loads of nutrient nitrogen for Switzerland

The SMB method was applied to forests, the empirical method to (semi-)natural ecosystems.



Critical loads of nitrogen for (semi-)natural ecosystems

The critical loads for natural and semi-natural ecosystems (CL_{emp}[N]), empirical method, chapter 2.2) are in the range from 4 to 15 kg N ha^{-1} a^{-1} covering a total sensitive area of 18 584 km². The most sensitive ecosystems are alpine lakes, raised bogs, Littorellion, (sub-)alpine scrub habitats and (sub-)alpine grassland (4–10 kg N kg N $ha^{-1}a^{-1}$). The largest part of these sensitive areas is covered by (sub-)alpine grassland and scrub habitats; these poorly managed pastures and meadows represent a very important part of the Alpine vegetation. Because of their short vegetation period, the low temperatures and the relatively poor soils, their natural nitrogen demand, and consequently their critical load, is low. Critical load between 10 and 15 kg N ha^{-1} a^{-1} were assigned to selected species-rich montane and sub-alpine grassland, fens and poorly managed forests with especially rich ground-flora.

In the Swiss Plateau, between the Alps and the Jura mountains, relatively few sensitive areas can be found on Figure 26. This can be explained by the fact that in this region a large part of the (semi-)natural ecosystems has disappeared, mainly due to intense agricultural management (e.g. mountain hay meadows, see Bosshard 2015). Two main reasons can be mentioned for the poor occurrence of (semi-)natural ecosystems in the Central and Southern Alps: (1) Acidic alpine grasslands such as *caricion curvulae / sempervirentis* and *nardion* have not been included, and (2) large areas south of the Alps are covered by forest, which was included in the SMB method.

Critical loads of nitrogen for forests

The critical loads for productive and managed forests (CL_{nut}(N), calculated with the SMB method (see chapter 2.3) are in the range from 10 to 25 kg N ha^{-1} a^{-1} covering a total sensitive area of 10 632 km² (Fig. 27). As described in Chapter 2.3.1, the lower range of 10 kg N ha^{-1} a^{-1} was set with intent to avoid implausible low values. The variation of the critical loads is mainly due to the removal of nitrogen by harvesting and denitrification losses. The average harvesting rates in the mountainous regions and south of the Alps are lower than in the Plateau (Fig. 12). The denitrification rates depend on the percolation conditions of the soils. The highest denitrification rates and

consequently the highest critical loads are found on wet soils of the lower Alps and in north-eastern Switzerland (Fig. 14).

4.2 Deposition of Nitrogen

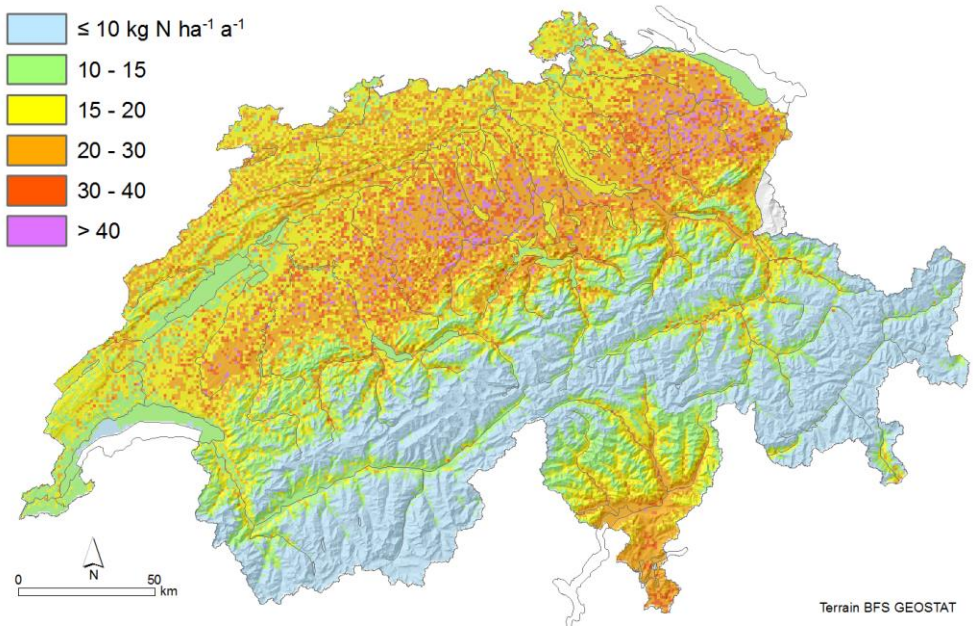
The modelled nitrogen depositions in 2010 are shown in Figure 30. In general, the depositions are higher in the lowlands and valleys, where the pollutant concentrations are higher. This is the case for NH_3 due to animal husbandry and for NO_2 due to intense traffic and settlements. The atmospheric deposition of gaseous NH_3 is strongly correlated with the spatial distribution of NH_3 emissions (see Chapter 3.5). Intense animal husbandry and subsequent high NH_3 emissions occur mainly in central and eastern Switzerland. The highest depositions are calculated on forest stands in these high emission regions. In mountainous regions the density of livestock and NH_3 emissions is much lower.

Total nitrogen deposition in Switzerland

As shown in Table 11, the resulting total nitrogen deposition in Switzerland for the year 2010 amounts to 66.7 kt N a^{-1} (16.3 kg ha^{-1} a^{-1}) to which gaseous NH_3 contributes 35%, wet NH_4^+ 26%, wet NO_3^- 18%, gaseous NO_2 10%, dry NH_4^+ 4%, gaseous HNO_3 3% and dry NO_3^- 2%. The share of reduced nitrogen compounds (NH_y-N) is 66%. The dry deposition rates depend on the type of land use. It is evident that forests get a larger load of dry deposition per unit of surface area compared with open land due to higher deposition velocities ("filtering effect", see chapters 3.3 and 3.4).

Fig. 30 > Total deposition of nitrogen calculated on a 1 x 1 km² raster

Year 2010.



Tab. 11 > Total deposition of N compounds in Switzerland

Year 2010. Units: kt N a^{-1} (italic: kg N ha^{-1} a^{-1}).

Receptors	Compounds	Forest 11 366 km²		Non-forest 29 683 km²		Total 41 049 km²	
Wet	NH_4^+	5.6	*5.0*	12.0	*4.1*	17.7	*4.3*
Dry	NH_4^+ aerosol	1.4	*1.2*	1.5	*0.5*	2.9	*0.7*
	NH_3 gas	10.1	*8.9*	13.5	*4.6*	23.6	*5.8*
Total NH_y-N		**17.2**	***15.1***	**27.1**	***9.1***	**44.2**	***10.8***
Wet	NO_3^-	4.0	*3.5*	8.3	*2.8*	12.3	*3.0*
Dry	NO_3^- aerosol	0.6	*0.5*	0.6	*0.2*	1.2	*0.3*
	NO_2 gas	3.4	*3.0*	3.5	*1.2*	6.9	*1.7*
	HNO_3 gas	0.6	*0.5*	1.5	*0.5*	2.1	*0.5*
Total NO_y-N		**8.6**	***7.6***	**13.9**	***4.7***	**22.5**	***5.5***
Total N		**25.8**	***22.7***	**41.0**	***13.8***	**66.7**	***16.3***

Nitrogen depositions resulting from different modelling approaches

The values in Table 11 can be compared with the independent results of the European EMEP-Model. EMEP applies an atmospheric transport model to the national emission data, resulting in a country-to-country transfer and deposition matrix with a spatial resolution of 50 x 50 km². The modelled total deposition for Switzerland in 2010 was 62 kt a^{-1} (Gauss et al. 2015), i.e. the difference is only -8%. Another independent model result was provided by Aksoyoglu (2014): She calculated dry N depositions in 2006 with the CAMx[13] model (raster size ~4 x 6 km²); the resulting total dry deposition for Switzerland was 32.7 kt N a^{-1} (difference -11%) and the gas deposition of NH_3 was 23.7 kt N a^{-1} (difference +0.2%).

An independent deposition study at a single site is available from 1992/1993: Neftel & Wanner (1994) carried out comprehensive field measurements on a meadow at Merenschwand (rural area of the Swiss Central Plateau) and found a total N deposition of 27–30 kg N ha^{-1} a^{-1} (depending on the measurement method for NH_3). This is in agreement with the temporally interpolated result of the deposition models 1990 and 2000 for the same site (28 kg N ha^{-1} a^{-1}).

The modelled depositions were also compared with throughfall measurements in forests (e.g. Thimonier et al. 2005). Generally, the total depositions derived from throughfall were lower than the modelled depositions. There are several possible reasons for this: (1) The model does not incorporate forest edge effects, which may overestimate dry nitrogen deposition at inner parts of forest areas, where the measurement sites are situated in general. (2) The N depositions derived from throughfall may underestimate direct canopy uptake of N. (3) Omitting stemflow in the measurements leads to underestimations of deposition. (4) At higher altitudes, snowfall can contribute to the underestimation of precipitation measurements. As mentioned in chapter 3.2, various parameters were adapted in the current model based on the study by Thimonier et al. (2005).

[13] www.camx.com

From 1990 to 2010 the total deposition in Switzerland was reduced by 23% (Table 12). 14% of this reduction took place between 1990 and 2000, i.e. the change was less pronounced since 2000, especially for the gaseous deposition of ammonia. This is in accordance with measured ammonia concentrations, which show no significant trend between 2000 and 2014 (Seitler & Thöni 2015). HNO_3 and aerosols contribute less than 10% to the total deposition. For simplification purposes and because of lack of data aerosols were left constant for the whole period; maps of HNO_3 were available for the years 1990 and 2000 and the latter was also used for 2010.

Tab. 12 > Total deposition von N compounds in Switzerland

Years 1990, 2000 and 2010. Units: kt N a^{-1}.

Compounds		1990	2000	2010
Wet	NH_4^+	24.9	21.4	17.7
Dry	NH_4^+ aerosol	2.9	2.9	2.9
	NH_3 gas	26.9	23.9	23.6
Total NH_y-N		**54.7**	**48.3**	**44.2**
Wet	NO_3^-	18.9	14.9	12.3
Dry	NO_3^- aerosol	1.2	1.2	1.2
	NO_2 gas	9.2	7.6	6.9
	HNO_3 gas	2.4	2.1	2.1
Total NO_y-N		**31.7**	**25.8**	**22.5**
Total N		**86.4**	**74.1**	**66.7**
Total N, percent		**100%**	**86%**	**77%**

4.3 Exceedances of Critical Loads of Nitrogen

Exceedances are calculated as deposition minus critical load. Thus, a negative exceedance means “not exceeded”. Exceedance of critical loads indicates a long-term risk of adverse effects in the ecosystems (see chapter 2.1) due to increased nitrogen deposition.

Spatial distribution of exceedances of critical loads

The spatial distributions of the exceedances of critical loads are displayed in Figure 31 ([semi-]natural ecosystems, empirical critical loads) and Figure 32 (forest, SMB method). The map in Figure 31 shows the exceedance of the most sensitive ecosystem type occurring in each 1 x 1 km² raster cell, i.e. the maximum exceedance.

The map in Figure 33 combines the exceedance of the critical loads for (semi-)natural ecosystems and forests showing the maximum exceedance per km².

Fig. 31 > Exceedance of critical loads for (semi-)natural ecosystems ($CL_{emp}[N]$) by nitrogen depositions in 2010

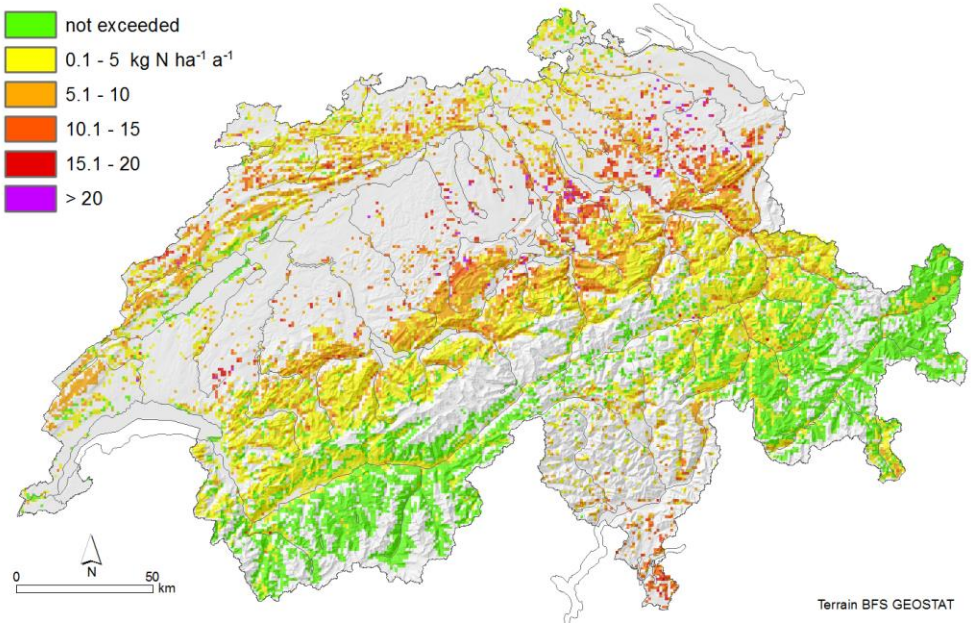
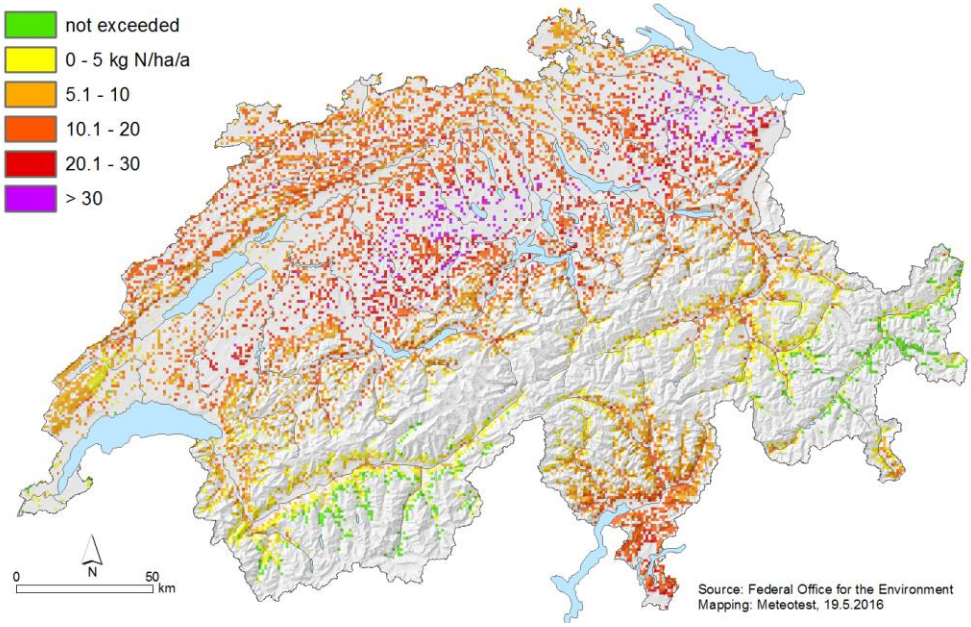
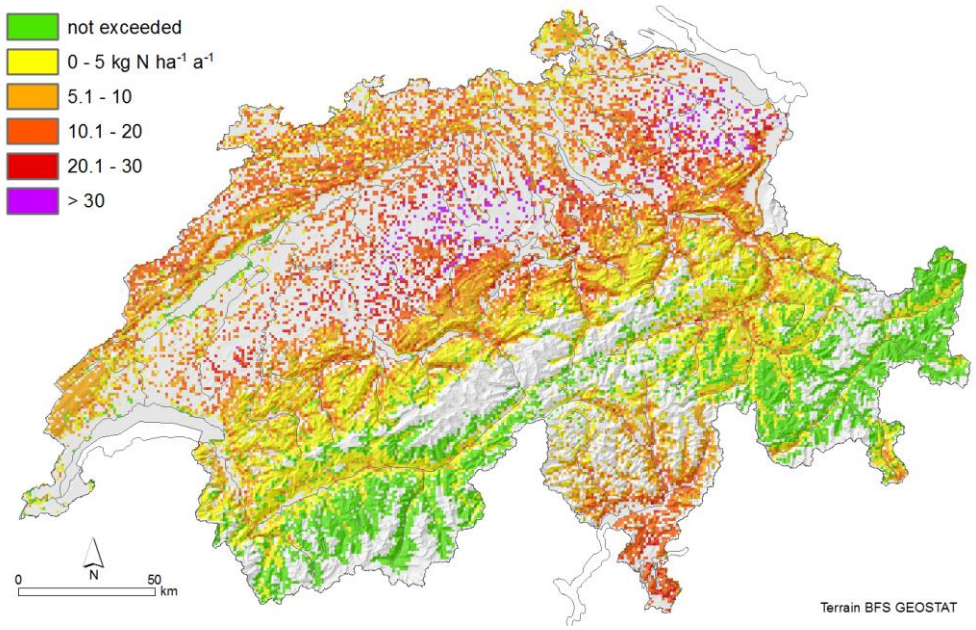


Fig. 32 > Exceedance of critical loads for productive forests ($CL_{nut}[N]$) by nitrogen depositions in 2010



Source forest grid: WSL 1990/92

Fig. 33 > Combined map showing the maximum exceedance of critical loads for forests (CL_{nut}[N]) and (semi-)natural ecosystems (CL_{emp}[N]) by nitrogen depositions in 2010 per km²



In 2010, the critical loads for natural and semi-natural ecosystems calculated with the empirical method are exceeded on 69% of the mapped area. The highest exceedances (more than 10 kg N ha^{-1} a^{-1}) occur in raised bogs and species-rich grassland in the lower Alps and in the Jura mountains, where low critical loads and relatively high depositions coincide. No exceedances or low exceedances (less than 5 kg N ha^{-1} a^{-1}) occur on species-rich grassland and alpine scrub habitats in the inner-alpine valleys, which are relatively dry regions (low wet deposition) and remote from the main sources of emissions. In the calculation of exceedances for forest ecosystems included in the empirical method (Table 2), deposition for open field conditions was used, because a part of these systems are open forests with a lower "filtering effect" in comparison with closed forests. Generally however, the exceedance in these unproductive forest ecosystems will be underestimated by such an approach.

The critical loads for productive forests calculated with the SMB method are exceeded on 95% of the mapped area (Figure 32). The exceedances are higher than those for the (semi-)natural ecosystems (Figure 31, the colour scheme in the two maps differs). This can be explained by the observation that the dry deposition is higher in forest stands than on open land, as the high surface roughness of forest canopies intensifies the dry deposition processes (chapters 3.3, 3.4, 4.2). The spatial distribution of the exceedances is determined to a large extent by present loads and not by critical loads. Again the inner-alpine valleys (Valais, Engadin) have the lowest exceedances.

Time series of exceedances of critical loads

Table 13 presents the history of exceeded area for the most important sensitive nature conservation areas in Switzerland (raised bogs, fens, dry grassland and forest); associated cumulative frequency distributions for the year 2010 are shown in Figure 34. The critical loads (according to Chapter 2.2.3), deposition and the resulting exceedance

were calculated on a 100 x 100 m² raster (cf. EKL 2014). For forests, the exceedance was calculated on the 1 x 1 km² sampling points of the national forest inventory as shown in Figure 32 (WSL 1990/92). For all these ecosystems a reduction of the exceeded area can be observed between 1990 and 2000 which continues between 2000 and 2010. However the percentage of exceeded area remains very high. The lowest exceedance was calculated for dry grassland (TWW) as these conservation areas are generally situated in dry regions and remote from strong sources of nitrogen emissions.

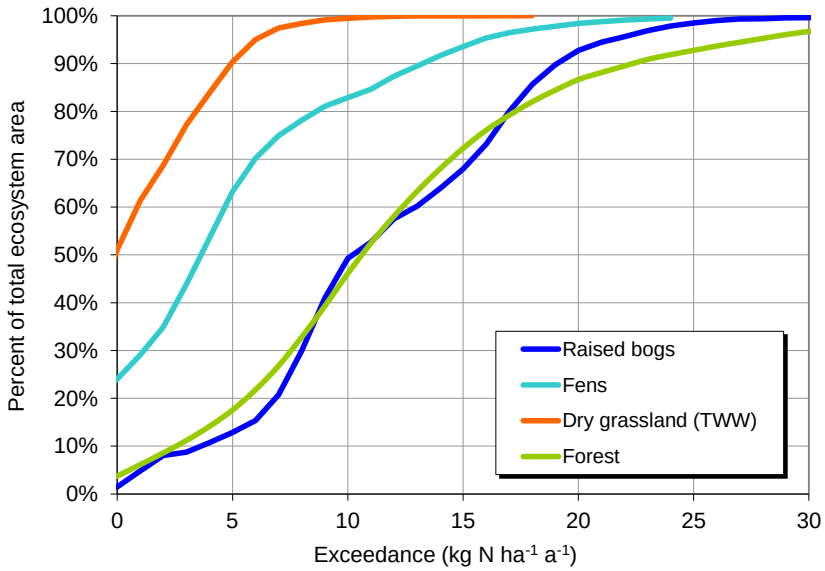
Tab. 13 > Exceedance of critical loads of nutrient nitrogen for different protected ecosystems in Switzerland in 1990, 2000 and 2010

Units: percent of total ecosystem area.

Ecosystem	Area km²	1990	2000	2010
Raised bogs	52	100	100	98
Fens	188	91	82	76
Dry grassland (TWW)	200	81	62	49
Forest	10 290	99	96	95

Fig. 34 > Cumulative frequency distribution of the exceedances of critical loads of nutrient nitrogen for different protected ecosystems

Units: percent of total ecosystem area. Nitrogen deposition: year 2010.



Adverse effects of exceedances of critical loads

The exceedances of critical loads shown in this Chapter indicate a risk of adverse effects in the long-term. Actually, harmful effects of excess nitrogen deposition were shown in various studies in Switzerland. E.g. for the raised bogs and fens, Klaus (2007) observed a declining quality of the ecosystem areas between surveys in 1997/2001 and 2002/2006, especially also an increase in nutrient availability. For beech forests, a clear decrease of root mycorrhiza with increasing N deposition was found (de Witte et al.

2015). Roth et al. (2013 and 2017) showed significant relations between the increase of N-deposition and decrease of species richness in mountain hay meadows and in (sub-) alpine scrub habitats (see also chapter 2.2.2) and Roth et al. (2015) detected negative relationships of N deposition and various biodiversity measures at the landscape scale.

4.4 Reliability of the Results

Uncertainties in the computation of critical loads and depositions involve associated uncertainty in the exceedance of critical loads. It is a challenging task to quantify that uncertainty as the relevant processes take place in open, interacting (eco)systems. As explicated by de Vries et al. (2015, chapter 25), the critical load approach meets precautionary principles to support environmental sustainability; it aims at "preventing damage caused by deposition even when scientific proof would not be conclusive". However, for supporting air pollution policies, information about the reliability of the critical load exceedances is needed. Therefore, a straight-forward sensitivity analysis was carried out and is presented in this chapter.

For empirical critical loads, the range of $CL_{emp}(N)$ given in the mapping manual partially reflects the uncertainty of the critical load (see chapter 2.2.1). E.g. for deciduous woodland the range is 10–20 kg N ha^{-1} a^{-1}. Thus, assuming a critical load value of 15 kg N ha^{-1} a^{-1}, the range corresponds to an uncertainty of approximately ±30%.

Uncertainty assessments of critical loads calculated with the SMB method ($CL_{SMB}[N]$) are quite rare (de Vries et al. 2015, chapter 25). For this sensitivity analysis the same uncertainty as for $CL_{emp}(N)$ is used (±30%). This assumption is also supported by a former sensitivity analysis of FOEFL (1996) where the influence of single input parameters to the SMB was analysed.

The error of modelled depositions can be quite large, e.g. for ammonia concentrations an error of 43% was calculated (chapter 3.5.3). However, the influence of these uncertainties on the reliability of the exceedance is limited when moving from site-specific calculations to regional applications as it was done here for the Swiss territory: As mentioned in chapter 4.2, the comparison of the sum of N deposition in Switzerland with two independent model results showed deviations of 8% (total deposition) and 11% (dry deposition).

Sensitivity analysis

Keeping these uncertainty ranges in mind, the following scenarios were defined in the sensitivity analysis:

- Standard run: These are the results presented in chapter 4.3 for the year 2010; those maps are considered as "best available" maps reflecting the current state of knowledge.
- CLN+30: All critical load values are increased by 30%.
- CLN-30: All critical load values are decreased by 30%.
- Dep-30: All deposition values are decreased by 30%.
- Dep+30: All deposition values are increased by 30%.

The outcome of the sensitivity analysis (expressed as percentage of exceeded ecosystem area) is shown in Table 14: For raised bogs and forests, the range of the exceedances is quite narrow in all scenarios (79%–100%), i.e. the magnitude of the exceedance is generally higher than the supposed uncertainties and therefore the reliability of the exceedance maps is very high. The range for dry grassland (TWW) is much wider (18%–83%), i.e. the prediction of exceedances is less reliable for this ecosystem. Nevertheless, a relevant part of the protected dry grasslands still exhibits an exceedance in scenario Dep-30 (minus 30% deposition). For fens, the range of exceedance is in-between (35%–92%).

Tab. 14 > Exceedance of critical loads of nutrient nitrogen for the standard run and four scenarios of the sensitivity analysis

Units: percent of total ecosystem area.

Ecosystem	CLN +30%	Standard 2010	CLN -30%
Raised bogs	92%	98%	100%
Fens	49%	76%	92%
Dry grassland (TWW)	26%	49%	83%
Forest	85%	95%	99%
Ecosystem	**Deposition -30%**	**Standard 2010**	**Deposition +30%**
Raised bogs	91%	98%	100%
Fens	35%	76%	90%
Dry grassland (TWW)	18%	49%	78%
Forest	79%	95%	98%

5 > Conclusions

This report provides an overall view of important aspects of the eutrophication problem in Switzerland. Maps of critical loads of nitrogen and of areas where they are exceeded were produced for whole Switzerland by combining data on the sensitivity of ecosystems and on atmospheric N deposition, consequently applying the critical load concept and the mapping methods proposed by the ICP Modelling & Mapping under the UNECE Convention on Long-range Transboundary Air Pollution (UNECE 2016). Since the last national report on critical loads (FOEFL 1996) the reliability of critical loads and modelled nitrogen depositions and critical load exceedances was improved, mainly by refining definitions and values of empirical critical loads under the Convention, by deriving country-specific critical loads based on Swiss biodiversity data, by enhancing the resolution of the deposition model and by improving the ammonia dispersion model.

The present atmospheric deposition of nitrogen in most forest sites and in many natural and semi-natural ecosystems in Switzerland exceeds the critical load with respect to eutrophication criteria. This means that the present nitrogen depositions do not meet the target of sustainability in large areas of Switzerland, as they are beyond the ecologically safe limit, above which adverse effects must be expected in the long-term. The situation is less severe for (sub-)alpine grassland, alpine scrub habitats and forests in the inner-alpine valleys (Valais, Grisons), where the critical loads are not exceeded or where the exceedances are relatively small.

While these conclusions can be drawn from the results of the study presented here, they are also supported by various studies based on field observations and experiments, especially concerning adverse effects on biodiversity and tree health. It is important to note that the natural deposition is estimated to be around 1–2 kg N ha^{-1} a^{-1} (Pardo et al. 2011, Bertills & Näsholm 2000, Binkley & Högberg 1997). Today such values can only be observed in very remote areas, e.g. northern Scandinavia.

This means that, as a consequence of anthropogenic emissions of nitrogen compounds, present loads of nitrogen in Switzerland are almost one order of magnitude higher than the natural nitrogen deposition. They range from 3 kg N ha^{-1} a^{-1} (1 percentile) to 44 kg N ha^{-1} a^{-1} (99 percentile) with an overall average of 16 kg N ha^{-1} a^{-1}.

In the long-term, the exceedances of critical loads indicate potential risks for forests such as changes in nutrient conditions, growth disturbances (unbalanced root/shoot ratios), increased susceptibility to gales, frost, drought, insect pests and pathogens, loss of stability and decreased biodiversity of the ground flora. For (semi-)natural ecosystems, exceedances indicate a potential risk for changes in competitive relationships between species, resulting in loss of biodiversity. The critical load approach does not allow quantifying those risks precisely, but one can say that they are potentially more severe the higher the exceedance is and the longer it lasts.

In order to improve the situation it is necessary to further reduce the emissions of air pollutants from agricultural activities (NH_3) and from combustion processes (NO_x). As the transboundary fluxes of nitrogen compounds are substantial, the control strategies and targets must be coordinated among the European countries: It will be crucial to achieve the targets set by the Gothenburg Protocol under the UNECE Convention on Long-range Transboundary Air Pollution until 2020 and to pursue further steps of effect-oriented international agreements beyond 2020. In addition, critical loads (and critical levels) can also be applied in local studies to assess compliance of projects in the context of national legislation, e.g. cantonal air pollution control action plans or environmental impact studies for agricultural and industrial facilities.

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Beat Rihm, December 2016

> Annex

Ammonia Dispersion Function

NH_3 concentration (1 m above ground, annual mean) as a function of the distance to a source (3 m above ground) emitting 1 kg NH_3 a^{-1} for distances between 50 m and 50 km (by Asman & Jaarsveld 1990):

Distance [m]	NH_3 concentration [µg m^{-3}]		Distance [m]	NH_3 concentration [µg m^{-3}]
50	4.97E-03		1 000	2.78E-05
60	3.96E-03		2 000	6.82E-06
70	3.23E-03		3 000	2.96E-06
80	2.68E-03		4 000	1.67E-06
90	2.25E-03		5 000	1.08E-06
100	1.92E-03		6 000	7.49E-07
120	1.44E-03		7 000	5.84E-07
140	1.12E-03		8 000	4.20E-07
160	8.89E-04		9 000	3.31E-07
180	7.25E-04		10 000	2.70E-07
200	6.03E-04		12 000	1.89E-07
250	4.03E-04		14 000	1.39E-07
300	2.88E-04		16 000	1.05E-07
350	2.16E-04		18 000	8.24E-08
400	1.67E-04		20 000	6.59E-08
450	1.33E-04		25 000	4.07E-08
500	1.09E-04		30 000	2.68E-08
600	7.65E-05		35 000	1.86E-08
700	5.65E-05		40 000	1.36E-08
800	4.34E-05		45 000	1.02E-08
900	3.43E-05		50 000	7.00E-09

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> Glossary

Ammonia
NH_3

Ammonium
NH_4^+

Base cations
Ca^{2+}, Mg^{2+}, K^+ and Na^+

Critical levels
"Concentrations of pollutants in the atmosphere above which adverse effects on receptors, such as human beings, plants, ecosystems or materials, may occur according to present knowledge" (UNECE 2004). Pollutants are in particular SO_2, NO_x (NO and NO_2), O_3 and NH_3.

Critical load
"A quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" (UNECE 2004).

Deposition
A quantitative estimate of atmospheric input of pollutants to specified elements of the environment.

Exceedance of critical loads
(Atmospheric) deposition minus critical load.

Load
Atmospheric deposition of chemical compounds. Units e.g. kg ha^{-1} a^{-1}.

Level
Concentration of chemical compounds in the atmosphere. Units e.g. µg m^{-3}.

Nitrate
NO_3^-

Nitric acid
HNO_3

Nitrogen dioxide
NO_2

Nitrogen saturation
Situation in an ecosystem where the availability of inorganic nitrogen is in excess of the total combined plant and microbial nutritional demand

> Abbreviations, figures and tables

Abbreviations

a
year

a.s.l.
above sea level

BDM
Biodiversity Monitoring Switzerland

CCE
Coordination Centre for Effects, RIVM, Bilthoven, The Netherlands

$CL_{emp}(N)$
Empirical critical loads of (nutrient) nitrogen

$CL_{nut}(N)$
Critical loads of nutrient nitrogen calculated with the SMB method

EMEP
Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

f_{de}
Denitrification fraction

FOEN
Federal Office for the Environment, Berne

FUB
Forschungsstelle für Umweltbeobachtung, Rapperswil, Switzerland

GVE
livestock unit corresponding to one milk cow (Grossvieheinheit)

ha
hectare

IAP
Institut für angewandte Pflanzenbiologie, Schönenbuch, Switzerland

ICP
International Cooperative Programme (under the LRTAP Convention)

ICP M&M
ICP Modelling and Mapping

kt
kiloton

LRTAP
(Convention on) Long-range Transboundary Air Pollution

eq
equivalents (=mol of charges)

NABEL
Nationales Beobachtungsnetz für Luftfremdstoffe (national air pollution monitoring network)

NFC
National Focal Centres for mapping activities under the Convention on LRTAP

NFI
National Forest Inventory (EAFV 1988)

NH_3
ammonia (gas)

NH_x
reduced nitrogen compounds: NH_3 and NH_4^+

NO_y
oxidised nitrogen compounds: NO_2, NO_3^- and HNO_3

N_i
long-term sustainable immobilization rate of N in soil organic matter

$N_{le(acc)}$
acceptable nitrogen leaching rate from the soil to the groundwater

N_u
net nitrogen uptake by vegetation

PM10
particulate matter (aerosols) with a diameter below 10 µm

SFSO
Swiss Federal Statistical Office

SMB
Simple Mass Balance, method for calculating critical loads of nutrient nitrogen and critical loads of acidity

TWW
Dry Grasslands of National Importance, National Inventory

UNECE
United Nations Economic Commission for Europe

WGE
Working Group on Effects (Convention on LRTAP)

WHO/Europe
World Health Organisation, Regional Office for Europe

WSL
Eidgenössische Forschungsanstalt für Wald, Schnee und Landschaft (Swiss Federal Institute for Forest, Snow and Landscape Research), Birmensdorf

µg
microgram

Figures

Tables