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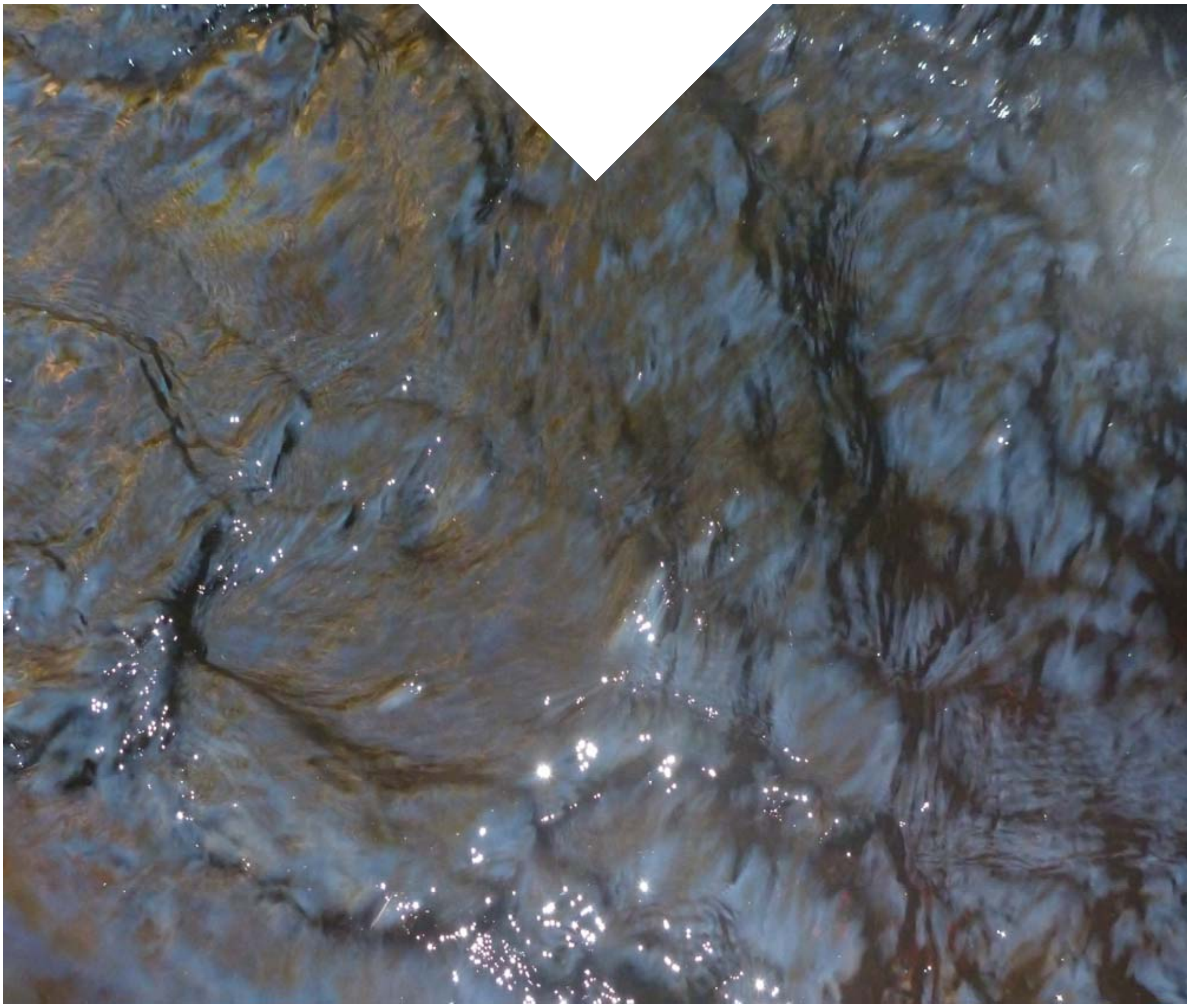
Critical limits for surface water acidification in Norwegian critical loads calculation and Water Framework Directive classification



Preface

In this report the critical limits for surface water acidification used in the UNECE Convention on Long-range Transboundary Air Pollution and the Water Framework Directive implementation in Norway have been compared. The objective was to investigate if there is consistency between requirements set by the two different management regulations.

The project is related to NIVA's role as National Focal Centre under the UNECE International Cooperative Programme on Modelling and Mapping of Critical Loads & Levels and Air Pollution Effects, Risks and Trends (ICP M&M).

The project has been led by Kari Austnes. Espen Lund has been responsible for calculations and map production.

This work has been conducted under contract 14078057 from the Norwegian Environment Agency. The contact person at the Norwegian Environment Agency was Gunnar Skotte.

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Kari Austnes

COLOPHON

Executive institution

Norwegian Institute for Water Research (NIVA)

Project manager for the contractor	Contact person in the Norwegian Environment Agency
Kari Austnes	Gunnar Skotte

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Author(s)

Kari Austnes and Espen Lund

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Grenseverdier for forsuring av overflatevann i beregning av tålegrenser og klassifisering etter vannforskriften i Norge.
Critical limits for surface water acidification in Norwegian critical loads calculation and Water Framework Directive classification.

Summary - sammendrag

This study compared the critical limits for surface water acidification used in the critical loads calculation under the UNECE Convention on Long-range Transboundary Air Pollution and the EU Water Framework Directive (WFD) classification in Norway. Critical loads based on the ANClimit,oaa,var from the critical loads work and the ANC good-moderate boundary values from the second WFD classification manual gave fairly harmonised results.
I denne studien er grenseverdier for forsuring av overflatevann som brukes ved beregning av tålegrenser under UNECEs konvensjon for langtransporterte luftforurensninger og i klassifisering etter vannforskriften i Norge sammenlignet. Tålegrenser basert på ANClimit,oaa,var fra tålegrensearbeidet og god/moderat-grensene for ANC i den andre klassifiseringsveilederen gav ganske like resultater.

4 emneord	4 subject words
Forsuring, grenseverdier, tålegrenser, Vannforskriften	Acidification, critical limits, critical loads, Water Framework Directive

Front page photo

Espen Lund

Norwegian Institute for Water Research
– an institute in the Environmental Research Alliance of Norway

REPORT

Main Office
Gaustadalléen 21
NO-0349 Oslo, Norway
Phone (47) 22 18 51 00
Telefax (47) 22 18 52 00
Internet: www.niva.no

NIVA Region South
Jon Lilletuns vei 3
NO-4879 Grimstad, Norway
Phone (47) 22 18 51 00
Telefax (47) 37 04 45 13

NIVA Region East
Sandvikaveien 59
NO-2312 Ottestad, Norway
Phone (47) 22 18 51 00
Telefax (47) 62 57 66 53

NIVA Region West
Thormøhlens gate 53 D
NO-5006 Bergen Norway
Phone (47) 22 18 51 00
Telefax (47) 55 31 22 14

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Abstract

The objective of this study was to compare the critical limits for surface water acidification used in the critical loads calculation under the UNECE Convention on Long-range Transboundary Air Pollution and the EU Water Framework Directive (WFD) classification in Norway. The critical limits to be compared were the $ANC_{limit,oaa,var}$ from the critical loads calculation and the ANC *good-moderate* boundary values from the WFD classification. They are not directly comparable, but were compared by using them to calculate critical loads. ANC *good-moderate* boundary values from the first WFD classification manual gave unrealistic results, showing that these boundary values were too high. Critical loads based on the $ANC_{limit,oaa,var}$ from the critical loads work and the ANC *good-moderate* boundary values from the second WFD classification manual gave fairly harmonised results. Thus, the requirements set by the two management systems are similar. However, the WFD requirements are somewhat less strict overall, and especially for regions with naturally low buffering capacity or high TOC concentration. Splitting the upper TOC category is recommended. Further harmonisation of the critical limits is possible, but the WFD concept of discrete boundary values sets limitations to complete harmonisation. Updating the $ANC_{limit,oaa,var}$ based on changes in TOC concentration could be considered.

4 keywords, Norwegian	4 keywords, English
1. Forsuring	1. Acidification
2. Grenseverdier	2. Critical limits
3. Tålegrenser	3. Critical loads
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Kari Austnes
Kari Austnes
Project Manager

Øyvind Kaste
Øyvind Kaste
Research Manager

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Content

Summary

There are two sets of political regulations used in management of surface water acidification in Norway: The UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) and the EU Water Framework Directive (WFD). Both use critical limits to define the chemical conditions necessary to achieve the desired biological conditions. The objective of this study was to compare the critical limits used in the LRTAP Convention and WFD implementation in Norway, as it is important that the same requirements are set across management systems.

The acid neutralising capacity (ANC) is used as chemical criterion in both the critical loads calculation under the LRTAP Convention and in the WFD classification. The critical limits to be compared were the $ANC_{limit,oaa,var}$ from the critical loads calculation and the ANC *good-moderate* boundary values from the WFD classification. Both vary with buffering capacity and humic content, giving higher critical limits at high buffering capacity and/or humic content. However, the way in which they vary with these factors makes it impossible to compare the critical limits directly. Hence, they were compared by using them to calculate critical loads.

Using ANC *good-moderate* boundary values from the first WFD classification manual to calculate critical loads gave unrealistic results, showing that the boundary values given here were too high. Critical loads based on the $ANC_{limit,oaa,var}$ from the critical loads work and the ANC *good-moderate* boundary values from the second WFD classification manual gave fairly harmonised results. Thus, the requirements set by the two management systems are similar.

However, discrepancies are found between the two systems. The WFD requirements are somewhat less strict overall. Especially for regions with naturally low buffering capacity, the requirements set by the $ANC_{limit,oaa,var}$ appear stricter. It is not possible to say which set of critical limits are more correct with respect to varying with different buffering capacity, though. At high TOC concentration the *good-moderate* boundary values seem to be insufficiently strict, and splitting the upper TOC category, giving a larger range of boundary values, is recommended. Moreover, the concept of discrete boundary values in the WFD introduces uncertainty at type boundaries.

Further harmonisation of the critical limits is possible, but the WFD concept of discrete boundary values sets limitations to complete harmonisation. Updating the $ANC_{limit,oaa,var}$ based on changes in TOC concentration could be considered. In general, however, the lack of more recent chemical and biological data limits the potential both for improvements and harmonisation of the critical limits.

Sammendrag

Det er to typer politiske reguleringer som brukes i forvaltningen knyttet til forsuring i ferskvann i Norge: UNECEs konvensjon for langtransporterte luftforurensninger (LRTAP) og EUs rammedirektiv for vann (Vannforskriften). Begge bruker grenseverdier for å definere de kjemiske forholdene som er nødvendige for å oppnå ønskelige biologiske forhold. Formålet med dette arbeidet var å sammenligne grenseverdiene brukt i LRTAP-konvensjonen og i implementeringen av vannforskriften i Norge, siden det er viktig at det settes de samme kravene på tvers av forvaltningssystemer.

Syrenøytraliserende kapasitet (ANC) brukes som kjemisk kriterium i både tålegrenseberegningene under LRTAP-konvensjonen og i klassifiseringen i henhold til vannforskriften. Grenseverdiene som skulle sammenlignes var $ANC_{limit,oaa,var}$ fra tålegrenseberegningene og god/moderat-grensene for ANC fra vannforskriftklassifiseringen. Begge varierer med bufferkapasitet og humusinnhold, grenseverdiene øker ved høy bufferkapasitet og/eller høyt humusinnhold. Måten de varierer med disse faktorene gjør imidlertid at det ikke går an å sammenligne grenseverdiene direkte. Derfor ble de sammenlignet ved å bruke dem til å beregne tålegrenser.

Når god/moderat-grensene for ANC fra den første klassifiseringsveilederen ble brukt til å beregne tålegrenser, ble resultatene urealistiske, som viser at disse god/moderat-grensene var for høye. Tålegrenser basert på $ANC_{limit,oaa,var}$ fra tålegrensearbeidet og god/moderat-grensene for ANC fra den andre klassifiseringsveilederen gav ganske like resultater. Kravene som stilles i de to forvaltningssystemene er dermed lignende.

Det ble likevel funnet avvik mellom systemene. Kravene i vannforskriften er gjennomgående noe mindre strenge. Særlig for områder med lav bufferkapasitet ser det ut til at $ANC_{limit,oaa,var}$ stiller strengere krav. Det er imidlertid ikke mulig å si hvilken type grenseverdi som på best måte tar hensyn til variasjon i bufferkapasitet. Ved høy TOC-konsentrasjon ser det ut til at god/moderat-grensen setter for milde krav, og det er anbefalt å dele den øvre TOC-kategorien, som vil gi et større spenn i grenseverdiene. Man ser også at konseptet med diskrete grenseverdier i vannforskriften fører til usikkerheter ved typegrensene.

Videre harmonisering av grenseverdiene er mulig, men konseptet med diskrete grenseverdier i vannforskriften setter begrensninger for fullstendig harmonisering. Det kan vurderes å oppdatere $ANC_{limit,oaa,var}$ basert på endringer I TOC-konsentrasjonen. Generelt legger imidlertid mangelen på nyere kjemiske og biologiske data begrensninger både for forbedringer og harmonisering av grenseverdiene.

1. Introduction

1.1 Objective

There are two sets of political regulations used in management of surface water acidification in Norway: The UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) and the EU Water Framework Directive (WFD). Both use critical limits to define the chemical conditions necessary to achieve the desired biological conditions. Setting such limits is not straightforward, and the limits have been subjects of discussion for many years. The objective of this study was to compare the critical limits used in the LRTAP Convention and WFD implementation in Norway, as it is important that the same requirements are set across management systems.

1.2 Critical loads and critical limits for acidification of surface waters

The atmospheric transport of acidifying compounds from industrial emissions and the subsequent deposition of such compounds, resulting in acidification of surface waters and negative effects on aquatic biota, led to the development of the concept of *critical loads for acidification of surface waters*. These critical loads quantify the acid deposition that an area can tolerate without negative effects on aquatic biota (often represented by brown trout). By comparing the critical loads with deposition data one can identify areas where critical loads are exceeded, and thus at risk of surface water acidification (Lund et al., 2012).

The concept of critical loads has been a scientific tool for assessing the problem of acidification and for the international work on reducing acidifying emissions in Europe and North America. It is a basis of both the sulphur protocol (1994) and the multi-pollutant protocol (1999) of the LRTAP Convention (http://www.unece.org/env/lrtap/status/lrtap_s.html).

Critical loads can be estimated using steady state models. The most commonly used are the Steady-State Water Chemistry (SSWC) model and the First-order Acidity Balance (FAB) model (Henriksen and Posch, 2001). Fundamental to both models is the critical limit of acid neutralising capacity (ANC), i.e. the ANC_{limit}. The critical limit is the link between surface water chemistry and biological response, and is set to avoid harmful effects on selected biota.

In the Norwegian calculations of critical loads, the ANC_{limit} was originally set to a constant, 20 µeq/l, based on surveys on fish in Norwegian lakes (Lien et al., 1996). This ANC_{limit} gives a 95% probability of no damage to fish populations. Later, the variable ANC_{limit} was introduced, based on the observation that for a given ANC there exist lakes of varying sensitivity. Conceptually, less sensitive systems should have a higher ANC_{limit} since they will generally have a higher biological diversity, which requires a higher ANC_{limit} to be held intact (Henriksen and Posch, 2001). The variable ANC_{limit} is denominated $ANC_{limit,var}$ and is mathematically described as

$$[ANC]_{limit,\ var} = k*Q*[BC^*]_0/(1+k*Q) \qquad (1)$$

where k is the proportionality constant describing the linear relationship between the $[ANC]_{limit}$ and the critical load, set to 0.25 yr/m, based on experience from the Nordic countries (for a critical load of 200 meq/m^2/yr the $[ANC]_{limit}$ should not exceed 50 meq/m^3), Q is the discharge and $[BC^*]_0$ is the sea-salt corrected pre-acidification base cation concentration. $ANC_{limit,\ var}$ has a range 0–50 µeq/l (if the expression gives a value higher than 50 µeq/l it is set to 50 µeq/l).

An additional adjustment to the ANC_{limit} was introduced to take into account the effect of naturally occurring organic acids (Lydersen et al., 2004). Many Norwegian lakes are humic, and part of the organic acids will act as strong acid anions. An adjusted ANC, taking this contribution into account, gave a slightly better fit with fish status. The organic acid adjusted ANC is expressed as

$$[ANC]_{oaa} = [ANC] - (1/3*m*[TOC]) \qquad (2)$$

where 1/3 expresses that one third of the organic acids will be negatively charged in most natural waters, m is the site density (set to 10.2 µeq/mg C, according to Hruska et al. (2001)) and [TOC] is the total organic carbon concentration in mg C/l. The $ANC_{limit,oaa}$ which gave a 95% probability of no damage to fish populations (brown trout) was 8 µeq/l. However, rather than using this value, a combination of the two approaches is used as critical limit in the current calculation of organically adjusted critical loads:

$$[ANC]_{limit,oaa,var} = k*Q*([BC^*]_0-1/3*m*[TOC])/(1+k*Q) \qquad (3)$$

The range is adjusted to -13-40 µeq/l and the k to 0.2 yr/m due to the general downwards adjustment caused by the organic acid adjustment (Hindar and Larssen, 2005).

1.3 WFD and boundary values for classification of surface waters with respect to acidification

To manage and protect European surface waters, the WFD[1] defines a classification system based on biological and physiochemical indicators (quality elements), where surface waters are assigned into five different classes of ecological status: *high*, *good*, *moderate*, *poor* or *bad*. In classes *high* and *good* no measures are needed, as waters in these classes are considered not to deviate or to deviate only slightly from the original conditions. Thus, the ecological status *good* is the environmental objective, and the *good-moderate* boundary is essential.

The physicochemical quality elements are supporting quality elements only, meaning that they are used only to adjust the conclusion from the biological quality elements if these classify the water body as *high* and *good*. Still, the *good-moderate* boundary of these quality

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[1] http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32000L0060

elements are important management targets, especially given that physicochemical quality elements are more frequently monitored than the biological quality elements.

The WFD is adopted into Norwegian legislation through Vannforskriften[2]. To support the implementation of the WFD, a classification manual has been produced: The first was produced in 2009 (Direktoratsgruppa Vanndirektivet, 2009) and a revised version was published in 2013 (Direktoratsgruppa Vanndirektivet, 2013). ANC is one of the physicochemical quality elements (along with pH and inorganic aluminium concentration) which is used to classify lakes and river water bodies with respect to acidification pressure. The *good-moderate* boundary for ANC reflects a physiochemical status that secures a functioning ecosystem and *good* status for the biological quality elements.

According to the principles of the WFD, different boundary values are given for different water body types. In the Norwegian classification of acidification status, the type factors considered relevant are calcium and humic content. The arguments for choosing these factors are much the same as the arguments for using the $\text{ANC}_{\text{limit,oaa,var}}$, i.e. a) that the biota is adapted to the natural ANC level, meaning that stricter requirements (higher ANC boundary values) are needed for high calcium lakes (parallel to high $[\text{BC}^*]_0$) (Direktoratsgruppa Vanndirektivet, 2013) and b) that higher ANC boundary values are needed with higher humic content, since part of the organic acids act as strong acid anions: Hesthagen et al. (2008) showed that at a given pH and inorganic aluminium concentration, the ANC boundary value must be higher in humic lakes.

In the first classification manual (Direktoratsgruppa Vanndirektivet, 2009), six sets of boundary values for lakes were given, based on two categories of calcium concentration (above or below 1 mg/l) and three categories of humic content (TOC concentration <2 mg/l, 2-5 mg/l and >5 mg/l). This gave a range in *good-moderate* boundary values of 20-40 µeq/l (Table 1). The *good-moderate* boundary values were primarily based on relationships between brown trout populations and ANC (Hesthagen et al. (2008); Solheim et al. (2008)).

In the second classification manual (Direktoratsgruppa Vanndirektivet, 2013) the lower calcium category was split in four, giving five categories of calcium concentration and a total of 15 sets of boundary values (Table 1). The reason to split the lower calcium category was the observed large variability in pre-acidification ANC, as modelled by the MAGIC model, especially below 1 mg Ca/l (Wright and Cosby, 2012). The MAGIC modelling also showed that pre-acidification ANC was frequently lower than the reference values given in the first classification manual, often even lower than the *good-moderate* boundary given. New reference values and *high-good* boundaries were identified from this modelling. The other boundary values were set based on new analyses on ANC-brown trout population relationships, splitting on the new calcium categories (Hesthagen, unpublished data), with subsequent downgrading to fit with the new reference values. The new *good-moderate* boundary values for ANC are in the range 0-30 µeq/l.

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[2] https://lovdata.no/dokument/SF/forskrift/2006-12-15-1446

Table 1. The ANC good-moderate (G/M) boundary values in the 2009 and 2013 WFD classification manuals for different lake types

Ca mg/l	TOC mg/l	G/M (µekv/l) 2009	G/M (µekv/l) 2013
<0.25	<2	20	0
0.25–0.5	<2	20	5
0.5–0.75	<2	20	10
0.75–1.0	<2	20	20
1–4	<2	20	20
<0.25	2–5	25	5
0.25–0.5	2–5	25	10
0.5–0.75	2–5	25	15
0.75–1.0	2–5	25	25
1–4	2–5	30	30
<0.25	>5	35	10
0.25–0.5	>5	35	15
0.5–0.75	>5	35	20
0.75–1.0	>5	35	30
1–4	>5	40	30

1.4 Comparing $ANC_{limit,oaa,var}$ and WFD *good-moderate* boundaries

Both the critical loads calculations and the WFD classification use ANC as chemical criterion for estimating status of surface waters with respect to acidification. ANC is the link between water chemistry and biological effects. To investigate consistency between the two management systems, the critical limits for ANC should be compared, i.e. the $ANC_{limit,oaa,var}$ for the critical loads calculation and the *good-moderate* boundary values for the WFD classification. However, the critical limits cannot be compared directly.

The ANC critical limits used in the critical loads calculations and the WFD classification are not fixed. They vary according to the same factors, i.e. buffering capacity (expressed as pre-acidification base cation concentration in the critical loads calculation and calcium concentration in the WFD classification) and humic content (expressed as TOC concentration), and they vary in the same way, i.e. the ANC critical limit increases with higher buffer capacity and humic content. However, whereas the *good-moderate* boundary values are discrete (for the 15 different type descriptions), the $ANC_{limit,oaa,var}$ are continuous (according to equation 3). Moreover, whereas different *good-moderate* boundary ANC values are given for different TOC concentration in the WFD classification, the effect of TOC concentration is integrated in the $ANC_{limit,oaa,var}$, i.e. it is a different definition of ANC, and cannot be directly compared with a normal ANC value.

Hence, to be able to compare the $ANC_{limit,oaa,var}$ and the *good-moderate* boundary values, both sets of values were used to calculate critical loads. In this way directly comparable numbers are produced, which are used to evaluate the consistency between the two types of management regulations.

2. Methods

2.1 Calculating critical loads

For this comparison, critical loads were calculated according to the Steady-State Water Chemistry (SSWC) model (Henriksen and Posch, 2001; UBA, 2004). According to the SSWC model, critical loads for acidification (CLA) are calculated as the pre-acidification flux of non-marine base cations (BC^*_0) minus a minimum ANC flux needed to protect biota (ANC_{limit}), i.e.

$$CLA = BC^*_0 - ANC_{limit} = Q*([BC^*]_0-[ANC]_{limit}) \qquad (4)$$

where Q is discharge.

When the $ANC_{limit,oaa,var}$ is applied, the following equation must be used, to adjust for the inclusion of the TOC-term in $ANC_{limit,oaa,var}$

$$CLA_{oaa} =Q*([BC^*]_0- [ANC]_{limit,oaa,var} -(1/3*m*[TOC])) \qquad (5)$$

where m is the site density (10.2 μeq/mg C). CLA_{oaa} just denotes that organic adjustment has been applied. The value is directly comparable with CLA in equation 4. For the SSWC model, the organic acid adjusted critical loads (CLA_{oaa}) were calculated for the whole country for the first time in the exceedance report from 2008 (Larssen et al., 2008), and in the 2012 report exceedances were only estimated based on CLA_{oaa} (Lund et al., 2012).

The critical loads for acidification in Norway are calculated for each grid cell in a grid of 2303 cells covering Norway (1/4 longitude and 1/8 latitude). Each grid cell is assigned a water chemistry based on the national lake surveys in 1986 and 1995, as described in the NFC report for Norway in Posch et al. (2012). Some cells have not been assigned a TOC concentration. In such cases it has been set to 1 mg/l (Larssen et al., 2008). The estimation of BC^*_0 follows the procedure of (Henriksen and Posch, 2001; UBA, 2004), with the updates described in Posch et al. (2012).

2.2 Critical loads using different critical limits

To compare the critical limits $ANC_{limit,oaa,var}$ and the *good-moderate* boundary values, critical loads were calculated by inserting $ANC_{limit,oaa,var}$ into equation 5 and the *good-moderate* boundary values as $[ANC]_{limit}$ into equation 4.

$ANC_{limit,oaa,var}$ has previously been estimated for each grid cell, using equation 3 and the assigned water chemistry. The calculated critical loads are called CLA_oaa. *Good-moderate* boundary values were defined for each grid cell based on the assigned calcium and TOC concentration for that grid cell, and the *good-moderate* boundary values given for the different lake types in the two classification manuals. Critical loads using *good-moderate* boundary values from the first classification manual as $[ANC]_{limit}$ are called CLA_WFD1, and when using the second classification manual they are called CLA_WFD2.

2.3 Calculating critical loads exceedance

Exceedance of critical loads was calculated for each grid cell as

$$Exceedance = S_{dep} + ENO_3 - CLA \qquad (6)$$

where S_{dep} is the sulphur deposition, in this report as given for the time period 2007-2011 (Aas et al., 2012) and ENO_3 is the nitrate output flux assigned to each grid cell. ENO_3 is used rather than the nitrogen deposition, as a portion of the deposited nitrogen is taken up in the ecosystems.

3. Results and discussion

3.1 Chemical variability and gradients

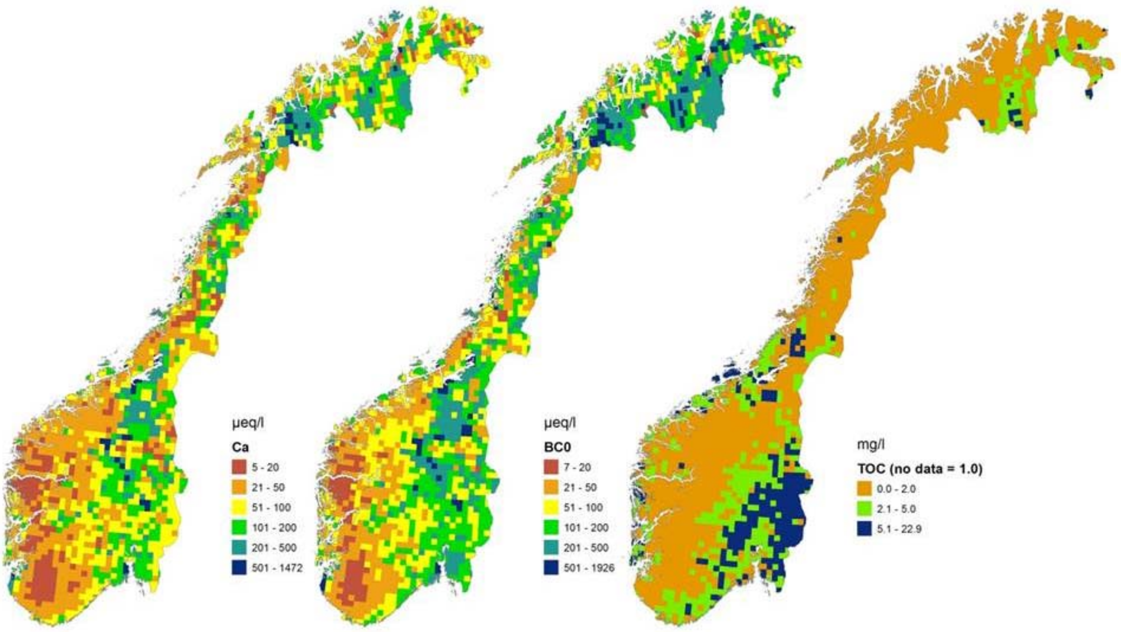

Figure 1. Calcium concentration (left), pre-acidification base cation concentration (BC^*_0: middle) and TOC concentration (right) across Norway.

Figure 1 shows the chemical variability across Norway for relevant parameters, as assigned to each grid cell (Ca and TOC concentration) or estimated from the assigned water chemistry (BC^*_0). The patterns in calcium concentration and BC^*_0 are quite similar, as they both reflect the buffering capacity in the catchments. The general pattern is lower buffering capacity in the western part of southern Norway and higher buffering capacity in the eastern part. In northern Norway there are more mixed conditions, but the majority of the area has higher buffering capacity. The differences in buffering capacity mainly reflect differences in geology.

The TOC concentration increases from the west to the east in southern Norway, and is also somewhat higher in mid-Norway and the northernmost county Finmark. The variability in TOC concentration mainly reflects differences in topography and land cover, with flatter areas with longer water residence times and more peatlands giving waters with higher TOC concentration.

3.2 ANC *good-moderate* boundary values

The ANC *good-moderate* boundary values assigned to the grid cells reflect the chemical variability (Figure 2). The *good-moderate* values are higher for grid cells with higher calcium and/or TOC concentration. Thus, independent on which classification manual is used, the ANC *good-moderate* boundary values are higher in the eastern part of southern Norway (with higher calcium and TOC concentrations) and lower in the western part. There are areas in mid- and northernmost Norway with higher *good-moderate* boundary values, but also areas in northern Norway with very low *good-moderate* boundary values, related to low calcium concentration.

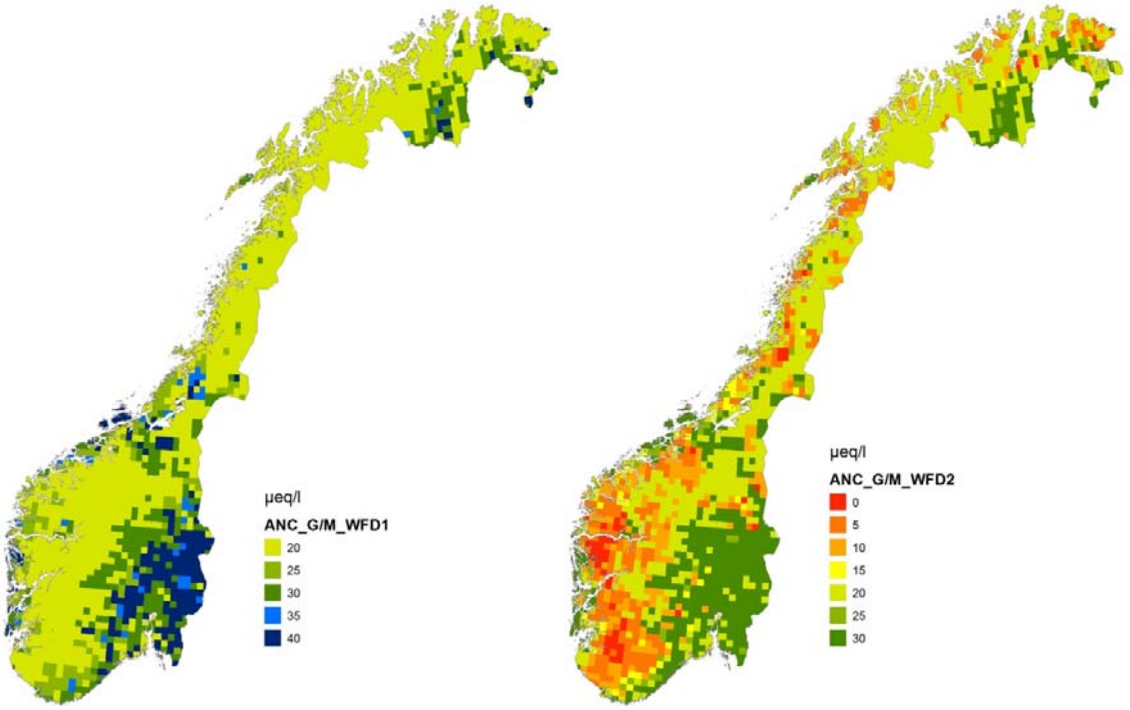

Figure 2. The ANC good-moderate boundary values based on the first classification manual (left) and the second classification manual (right)

Although the relative patterns are similar, the absolute values of the ANC *good-moderate* boundary values differ markedly depending on which version of the manual is used. Generally, the values are lower using the second classification manual. There is also larger differentiation in the region with lower calcium concentration (the western part of southern Norway). These effects were to be expected from the changes made from the first to the second classification manual (cf. section 1.3).

3.3 Comparing critical limits through critical loads and exceedances

As explained in section 1.4, the ANC$_{\text{limit,oaa,var}}$ and the ANC *good-moderate* boundary values cannot be compared directly. Hence, critical loads based on the three different sets of critical limits were calculated (Figure 3). First of all the critical loads are much higher when using the *good-moderate* boundary values from the second classification manual (CLA_WFD2), compared to the first (CLA_WFD1). Using the first manual the critical loads are negative in some regions, which clearly indicates that the *good-moderate* boundary values are too high. This happens in particular in the regions with low buffering capacity. This confirms that the boundary values in the first classification manual did not take sufficiently into account that some waters have naturally very low buffering capacity. The ANC *good-moderate* boundary values in the first classification manual will hence not be further discussed.

CLA_WFD2 are quite similar to the critical loads calculated based on the ANC$_{\text{limit,oaa,var}}$ (CLA_oaa). This confirms that the revisions made to the second classification manual were reasonable. The fact that the critical loads based on critical limits derived at least partly from different processes and calculations are quite similar, confirms the validity of these critical limits. So, in general the comparison shows good coherence between the critical limits, and at large they set similar requirements.

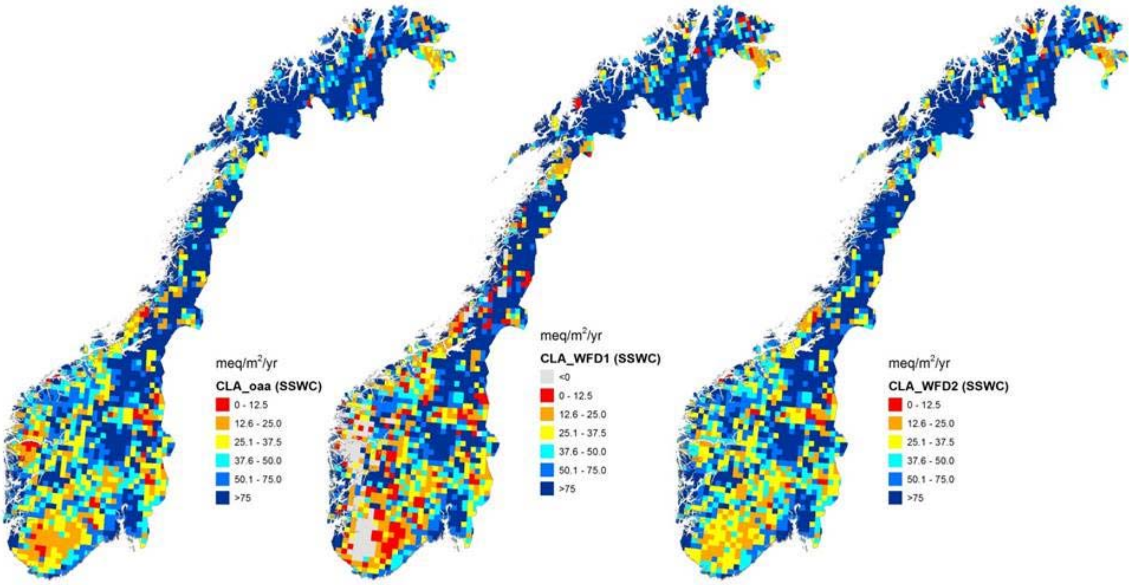

Figure 3. Critical loads calculated according to the SSWC model, using the ANC$_{\text{limit,oaa,var}}$ (left) or good-moderate boundary values as critical limits (middle: first classification manual; right: second classification manual).

However, there are differences between CLA_WFD2 and CLA_oaa, although not so easy to see from Figure 3. Figure 4 shows the exceedance based on the two sets of critical limits. The exceedance is slightly higher using CLA_oaa, meaning that the $ANC_{limit,oaa,var}$ set somewhat higher requirements than the WFD2 *good-moderate* boundary values. This is particularly evident for the western part of southern Norway, where there are not only larger areas with exceedance, but the exceedance is also higher.

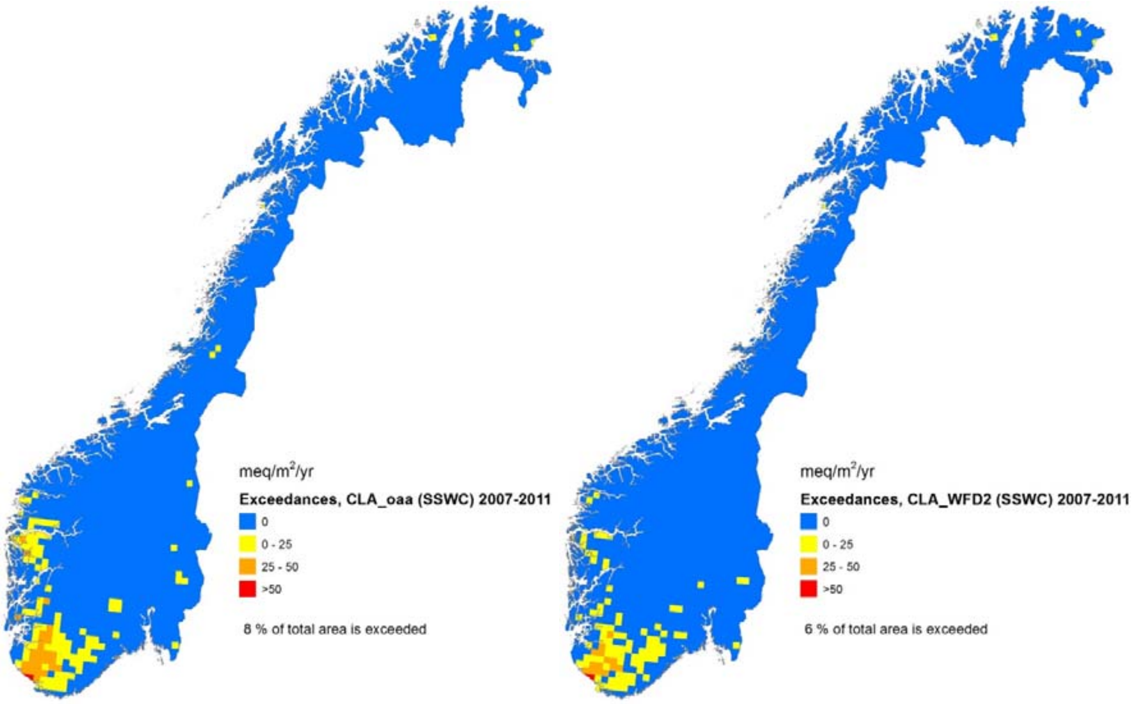

Figure 4. Critical loads exceedance for the period 2007-2011, calculated using CLA_oaa (left) and CLA_WFD2 (right).

Another way to identify the differences between the two sets of critical loads is to subtract one from the other. This is done in Figure 5. However, grid cells are left blank where both types of critical loads were higher than 90 meq/m^2/yr. In such areas there is no risk of acidification, and differences between the critical loads estimates are not relevant and only cause noise in the analysis. Figure 5 shows that in some regions, the CLA_oaa are highest (e.g. the middle part of southern Norway) and in some regions the CLA_WFD2 are highest (especially the western and easternmost parts of southern Norway). However, the differences are generally largest where CLA_WFD2 are highest, i.e. showing again that the WFD2 *good-moderate* boundary values set somewhat less strict requirements than the $ANC_{limit,oaa,var}$.

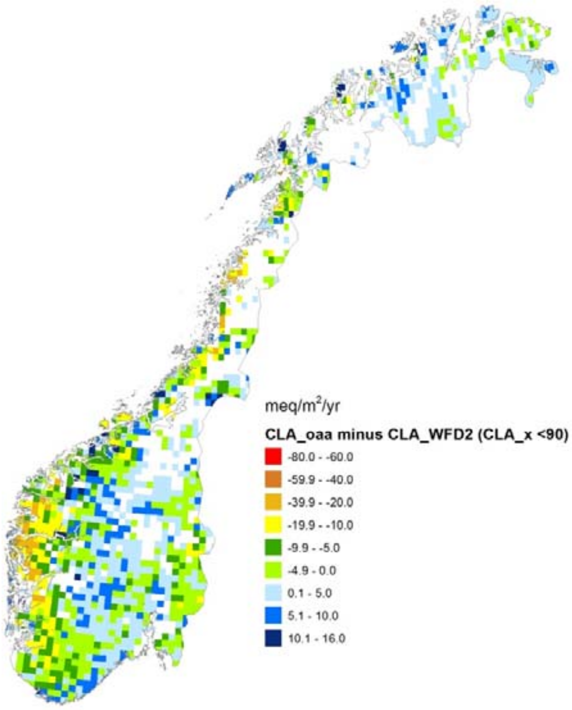

Figure 5. Difference between the critical loads estimates CLA_oaa and CLA_WFD2. Positive values (blue) means that CLA_oaa is higher than CLA_WFD2. Only grid cells where both CLAs are <90 meq/m^2/yr are coloured.

3.4 The effect of humic content and buffering capacity

The geographical variability in deviation between CLA_oaa and CLA_WFD2 is related to the different ways in which the critical limits vary with buffering capacity and humic content. It is difficult to disentangle to which extent the deviations are affected by the two different factors, i.e. whether it is the dependency of the critical limits on humic content or on buffering capacity which is the main source of deviation between the critical loads estimates. This is particularly because the two factors vary in the same manner, i.e. they are both low in the western part of southern Norway and both high in the easternmost part of southern Norway, the two regions with the largest deviations. However, looking at the two factors independently can give some indication.

In Figure 6 the difference between CLA_oaa and CLA_WFD2 is plotted against TOC concentration. This shows that at low TOC concentration there is a wide range in deviation, and it is both positive and negative, meaning that the dependency of the critical limits on the humic content is probably less important in this TOC concentration range. However, at TOC concentration > 5 mg/l, the deviation is mainly negative (i.e. CLA_WFD2 is highest), it is always negative at TOC concentration > 8 mg/l, and it becomes more negative as the TOC concentration increases further. This can explain the high deviation observed in easternmost Norway, where the TOC concentration is high. It may also indicate that the CLA_WFD2 (and hence the WFD2 *good-moderate* boundary) is not strict enough at high TOC concentration. This can be a result of the lack of differentiation between TOC categories above 5 mg/l. Whereas the $ANC_{limit,oaa,var}$ gradually increases as TOC increases, the *good-moderate* boundary remains the same above 5 mg/l (as long as the calcium concentration category is the same). This can give too low values in the upper TOC concentration range, as the boundary values are representative for an average TOC concentration within the category.

Figure 6 also shows that the deviation pattern shifts abruptly at the TOC category boundaries. Again, this is an effect of the discrete *good-moderate* boundary values, in contrast to the continuous $ANC_{limit,oaa,var}$. This creates artificially abrupt changes in CLA_WFD2 at the category boundaries.

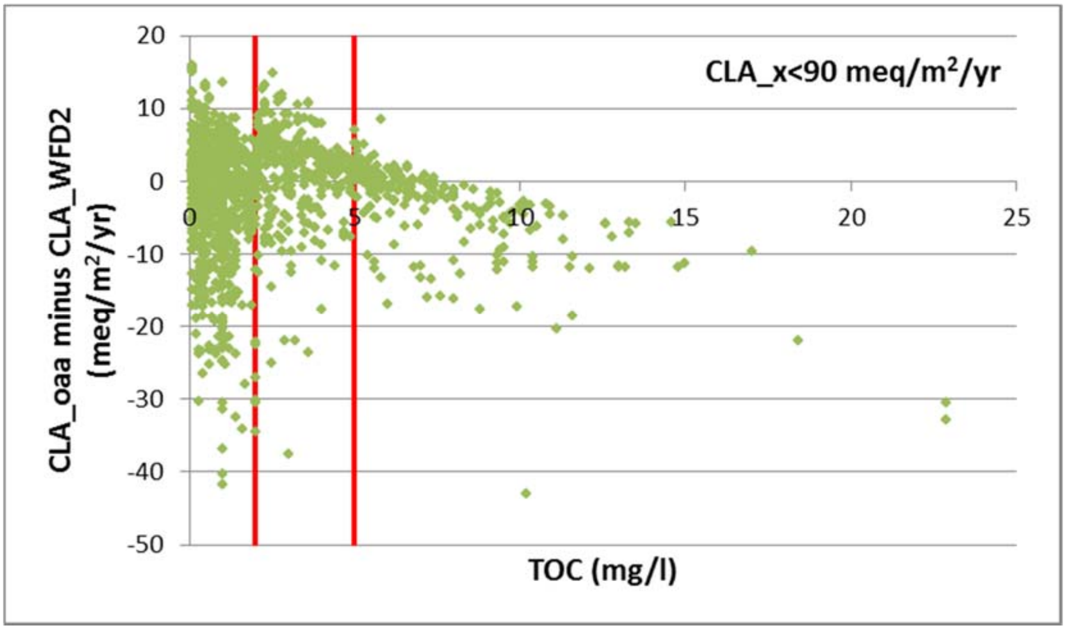

Figure 6. The relationship between the difference between CLA_oaa and CLA_WFD2 and TOC concentration. Only grid values where both CLAs are <90 meq/m²/yr are included. TOC category boundaries (WFD) are marked by red vertical lines.

Figure 7 shows the deviation in critical loads plotted against calcium concentration. Here as well one can see marked changes at calcium category boundaries. Transferred to the use of boundary values in WFD classification, it means that the classification is more uncertain for lakes close to the calcium category boundaries.

The largest positive deviations (CLA_WFD2 is lowest) are found in the region 0.75-1.25 mg/l calcium. Negative deviations are found along the whole range, but in particular below 0.75 mg/l calcium (37 µeq/l). These are mainly grid cells in the western part of southern Norway. So, there is clearly a differentiation between the two types of critical limits at very low buffering capacity. However, it is not possible to state from these data whether the $ANC_{limit,oaa,var}$ is too strict (giving too low critical loads) or the WFD good-moderate boundary values are too mild. The large negative deviations found at high calcium concentrations are for grid cells with high TOC concentration, so the deviations are not related to the calcium concentration. The patterns seen in Figure 7 were very much the same if plotting critical loads deviations against BC^*_0.

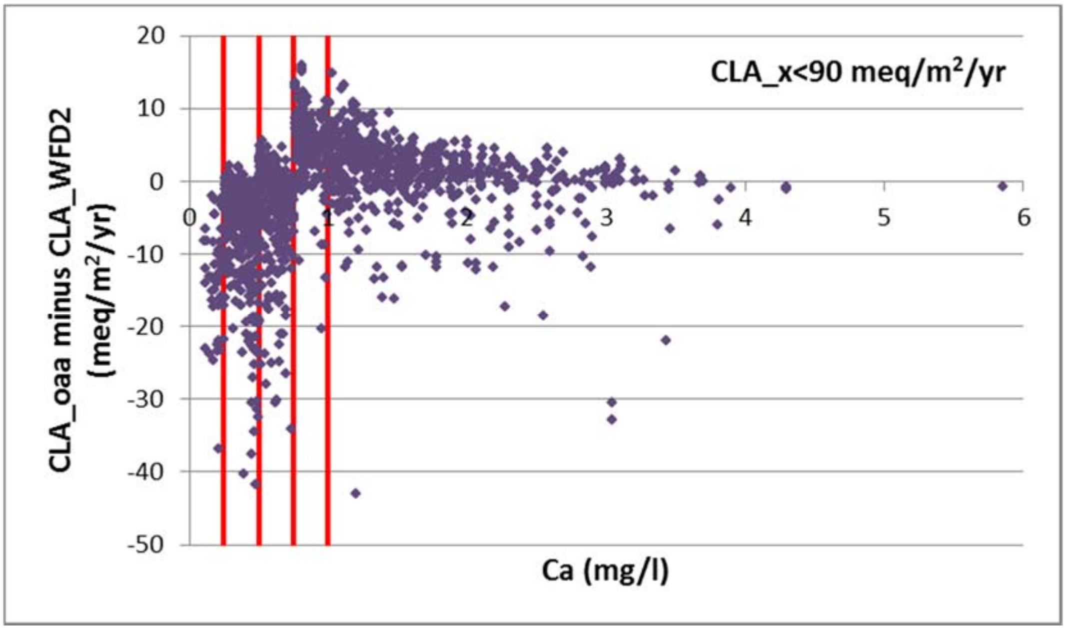

Figure 7. The relationship between the difference between CLA_oaa and CLA_WFD2 and calcium concentration. Only grid values where both CLAs are <90 meq/m^2/yr are included. Calcium category boundaries (WFD) are marked by red vertical lines.

So to summarise, CLA_oaa is stricter (lower) at very low calcium concentration while CLA_WFD2 is stricter at calcium concentration around or just below 1 mg/l. Differences in the influence of humic content is mainly found at high TOC concentration, where CLA_oaa is stricter. These effects are also seen when looking at differences in exceedance. In Figure 8 the exceedance for grid cells with exceedance only for one of the types of critical loads are plotted against calcium or TOC concentrations. For calcium concentration there is a clear shift around 0.8 mg/l, where grid cells with exceedance only for CLA_WFD2 are only found above this threshold. The same grid cells are found in the whole lower range of TOC concentration, but never above TOC concentration 7 mg/l.

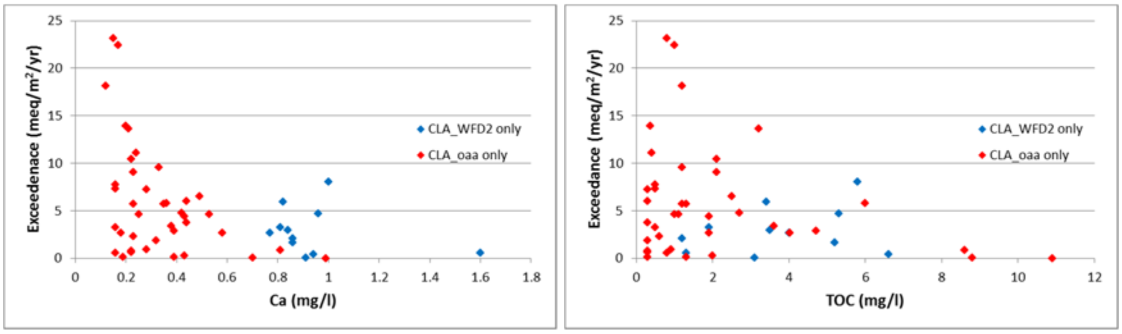

Figure 8. Exceedance for grid cells with exceedance only for one of the critical loads types (CLA_WFD2 or CLA_oaa) plotted against Ca concentration (left) or TOC concentration (right).

3.5 Harmonisation, limitations and possible improvements

Generally the critical limits for ANC in the two systems are fairly harmonised and much more in line after the revision of the classification manual. The critical limits used have been developed at different times, based on different data. Wright (2013) argued that differences in critical limits may be related to differences in timing of chemical and biological effects. Data from 1986 (used by Lien et al. (1986), Henriksen and Posch (2001) and Lydersen et al (2004)) represent a situation where chemical conditions were poor, but the negative biological response was still not at the maximum. Data from 1995 (used in the WFD classification manuals, but adjusted in the second version) represent a situation where there was chemical recovery, but very little biological recovery. Hence, the ANC boundary values based on 1995 data are likely to be too high, while ANC limits based on 1986 data may be too low. These considerations were taken into account in the second WFD classification manual, adjusting the critical limits found from the brown trout-ANC relationships downwards. This involved some expert judgement, drawing on both the critical loads work and dynamic modelling exercises like Wright and Cosby (2012). However, the $ANC_{limit,oaa,var}$ is even less directly based on biota-ANC relationships. The empirical relationships are only embedded in the range set and in the constant k. As such it is difficult to state which set of critical limits are more or less correct, although the relatively high degree of coherence confirm that both sets of critical values are reasonable. However, the discrepancy especially at low buffering capacity underlines the difficulty in setting critical limits. Using data collected in a dynamic situation adds to the difficulty. In the current further stage of recovery, new combined chemical and biological data sets could have been used to improve the critical limit estimates, if available.

Ideally there should be complete harmonisation, using one type of critical limits only when evaluating the effect of one type of pressure, in this case acidification. However, the WFD system requires discrete critical limits, i.e. boundary values given for distinct water types. As shown this is a clear disadvantage, in that the boundary values will be more correct for part of the TOC or calcium concentration range within a category, and it creates large uncertainties in classification of lakes close to the type boundaries. It was also shown that the number of TOC categories is too low, and that the > 5 mg/l category should be split. So – using the WFD boundary values directly in the critical loads calculations is not a good option, while using the $ANC_{limit,oaa,var}$ in the WFD classification is not allowed. However, the WFD boundary values have the advantage that they to a larger extent are based on empirical data than the $ANC_{limit,oaa,var}$, in particular the variability related to buffering capacity. A further harmonisation could be investigated, e.g. making the ANC_{limit} vary with calcium concentration (preferably the pre-acidification calcium concentration) in a similar, but smoothed (continuous) manner to the WFD *good-moderate* boundaries.

Another issue to consider in potentially improving the critical limits used in the critical loads calculations is the observed increasing TOC concentration (e.g. Monteith et al., 2008). In the WFD concept the boundary values are in principle dynamic, i.e. if the TOC concentration increases (sufficiently), the lake type changes and new boundary values apply. The critical loads are in principle constant, i.e. they are based on estimated pre-acidification chemistry (in practice they are revised as calculations and data improve). However, including the TOC in the $ANC_{limit,oaa,var}$ introduces a non-constant factor. The $ANC_{limit,oaa,var}$ should ideally be updated as the TOC concentration changes. A rough test was made, adjusting the $ANC_{limit,oaa,var}$ using relative changes in TOC concentration based on monitoring data from Norwegian lakes (Garmo et al., 2013). The effects on critical loads and exceedances were relatively small (an increase from 8 to 10% exceedance), but not negligible. The changes were largest in the regions with already higher TOC concentration. Updating the $ANC_{limit,oaa,var}$ based on changes in TOC concentration can be considered, but it requires a more sophisticated approach and preferably new data.

4. Conclusions

Critical limits for surface water acidification in Norwegian critical loads calculation and Water Framework Directive classification have been compared by applying them in critical loads calculations. Using ANC *good-moderate* boundary values from the first WFD classification manual to calculate critical loads gave unrealistic results, showing that the boundary values given here were too high. Critical loads based on the $ANC_{limit,oaa,var}$ from the critical loads work and the ANC *good-moderate* boundary values from the second WFD classification manual gave fairly harmonised results. Thus, the requirements set by the two management systems are similar.

However, discrepancies are found between the two systems. The WFD requirements are somewhat less strict overall. Especially for regions with naturally low buffering capacity, the requirements set by the $ANC_{limit,oaa,var}$ appear stricter. It is not possible to say which set of critical limits are more correct with respect to varying with different buffering capacity, though. At high TOC concentration the *good-moderate* boundary values seem to be insufficiently strict, and splitting the upper TOC category, giving a larger range of boundary values, is recommended. Moreover, the concept of discrete boundary values in the WFD introduces uncertainty at type boundaries.

Further harmonisation of the critical limits is possible, but the WFD concept of discrete boundary values sets limitations to complete harmonisation. Updating the $ANC_{limit,oaa,var}$ based on changes in TOC concentration could be considered. In general, however, the lack of more recent chemical and biological data limits the potential both for improvements and harmonisation of the critical limits.

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Norwegian Environment Agency
Telephone: +47 73 58 05 00 | Fax: +47 73 58 05 01
E-mail: post@miljodir.no
Web: www.environmentagency.no
Postal address: Postboks 5672 Sluppen, N-7485 Trondheim
Visiting address Trondheim: Brattørkaia 15, 7010 Trondheim
Visiting address Oslo: Strømsveien 96, 0602 Oslo

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