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Critical loads for lead (Pb) and cadmium (Cd) in surface waters in Norway

- evaluation of methodology and preliminary maps



Fagrapport nr. 119



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<p>Abstract</p> <p>Critical loads for lead (Pb) and cadmium (Cd) are calculated for Norwegian surface waters based on currently available methodology from the UNECE ICP Mapping and Modelling program. Two different values are used for critical limits. Using the critical limits recommended in the Mapping Manual the critical loads are high, well above the current deposition in Norway. Using critical limits from the lowest part of the range suggested, however, the critical loads are of similar size as current deposition. We also calculated the so called stand still loads, where the criteria is no increase in current concentration rather than a biological effect. The stand still loads are very low, for Cd in many cases 0, and usually lower than the current deposition.</p>

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Fagrapport nr. 119

**Critical loads for lead (Pb) and cadmium (Cd) in
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methodology and preliminary maps**

Preface

NIVA is the Norwegian Focal Centre in the UN-ECE ICP Mapping and Modelling programme and actively participates with data and methodology regarding critical loads, in particular for acidification. In Europe efforts are made to apply the effects-based critical loads approach also for heavy metals, and procedures for calculation for critical loads for lead and cadmium are available. In this report we present results of critical load calculations for lead and cadmium in Norwegian surface waters. The Norwegian Pollution Control Authority (SFT) has funded the work. Contact at SFT has been Tor Johannessen.

Oslo, February 2004

Thorjørn Larssen

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Summary

Critical loads for lead (Pb) and cadmium (Cd) are calculated for Norwegian surface waters based on currently available methodology from the UNECE ICP Mapping and Modelling program. Two different values are used for critical limits. Using the critical limits recommended in the Mapping Manual the critical loads are high, well above the current deposition in Norway. Using critical limits from the lowest part of the range suggested, however, the critical loads are of similar size as current deposition. We also calculated the so called stand still loads, where the criteria is no increase in current concentration rather than a biological effect. The stand still loads are very low, for Cd in many cases 0, and usually lower than the current deposition. The importance of weathering and in-lake retention appear to be small in the suggested model.

1. Background

Under the United Nations Economic Commission for Europe (UNECE) there are initiatives to test an effects-based approach as a possible tool in international agreements/negotiations to reduce emissions of heavy metals to the atmosphere. The International Cooperative Programme on Modelling and Mapping (ICP M&M) has developed a draft manual for application of a simple steady-state mass balance models for terrestrial and aquatic ecosystems (UBA 2003, de Vries et al. 1998). A "call for data" was submitted from the Coordination Center for Effects (CCE) late 2001, with deadline 2002. Norway did not submit data to the call. The results from the call, including preliminary maps, were presented in a report from the CCE (Hettelingh et al. 2002). Several countries submitted data for different terrestrial ecosystems, but country submitted data for aquatic ecosystems.

In addition to the effects-based critical loads approach, another method, called the stand-still approach, has also been suggested. The stand-still approach is not based on estimates of effects, but on the criterium that the concentration in the environment should not increase above present level. The stand-still loads are not currently in the scope of the ICP M&M since they are not effects-based; however, they may still be relevant, for instance as an intermediate step towards a dynamic modelling approach (Pers. comm. Gudrun Schütze).

In the present report we have used the suggested methodology to calculate effects-based critical loads and stand-still loads for lead and cadmium for Norwegian surface waters. We also illustrate the importance of some of the input parameters and the choice of values for the critical limits. The maps are preliminary only.

2. Lake survey data and measured concentrations

The data used in this report are from the Norwegian part of the dataset in the Nordic Lake survey 1995 (Skjelkvåle et al. 2001). The survey was based on a statistical selection of lakes, but with stratification related to lake size and north-south gradient. There are about 39,000 lakes in Norway $> 0.04 \text{ km}^2$ (4ha). Of these 2.5% (985 lakes) were sampled.

The results from the lake survey in 1995 showed that heavy metals measured in nearly 3000 Nordic lakes in general show low concentrations and distinct geographical patterns. The levels and distribution of heavy metals in Nordic lakes are mainly related to anthropogenic sources, natural sources in the catchment and water chemical conditions.

Of the anthropogenic sources, long-range transported air pollutants deposited both directly on the lake and indirectly on the catchment contribute to the large-scale concentration patterns several heavy metals in the Nordic lakes.

Soil- and surface-water pH and the concentration of total organic carbon (TOC) are important for the mobilisation of metals from the catchment to the runoff water. In particular TOC is an important factor and explains the large-scale variation in heavy metal concentration in lakes. About 40% of the total variance in the data material could be explained by variance in TOC and pH (where TOC is the most important of the two).

With some exceptions, the content of TOC in the waters is more important for controlling heavy metal concentration in lakes than the mineralogy of the bedrock and overburden. Also the influence of long-range transport is more important for the regional patterns for some of the heavy metals than the mineralogy of the bedrock and overburden on the Nordic scale.

Direct and indirect deposition of long-range transported air pollution is the most important factor for distribution of Pb, Cd, Zn and also to a certain degree Co. TOC-levels in lakes are important for Fe and Mn and also to a certain degree As, Cr and V. Bedrock geology is the major controlling factor for Cu and Ni and is also an important factor for controlling the concentration levels of As, Co Cr and V.

The results from the Nordic lake survey indicate that heavy metal pollution in lakes is a minor ecological problem on a regional scale in the Nordic countries. Certain areas, however, in particular in southern Norway and Sweden, are affected by long-range transport leading to increased concentrations of Pb, Cd and Zn in lakes above limits set by environmental authorities in Norway and Sweden. Since aquatic biota in these areas already is stressed due to effects of acid rain, this may be an important environmental problem in such areas.

Hg is not included in the Nordic Lake Survey, but other work has demonstrated that the Hg concentrations in fish has increased substantially, and that this is a large environmental problem.

Lead

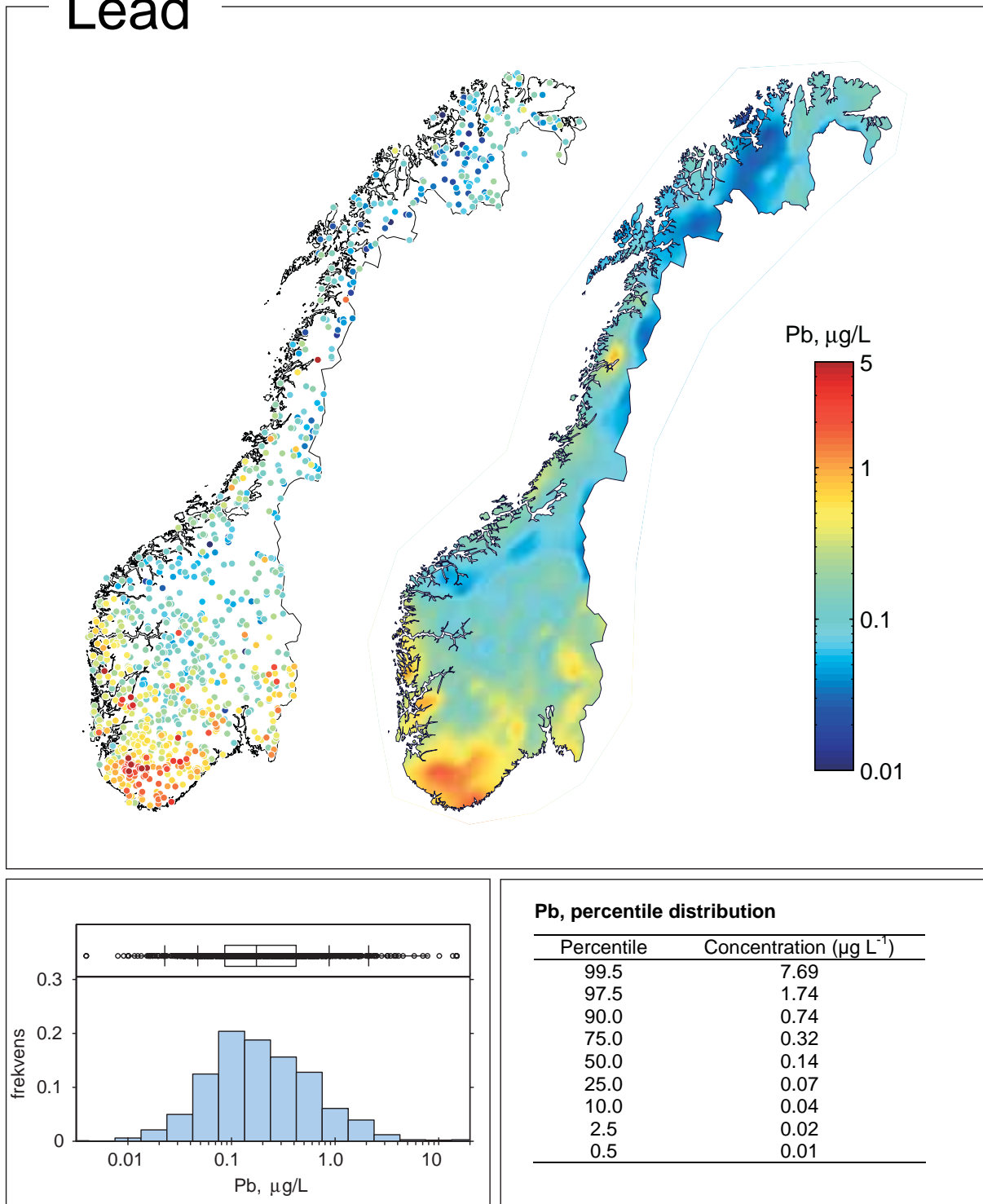


Figure 1. Concentrations of Pb in Norwegian lakes, autumn 1995. The map on the right side is based on spatial interpolation (kriging). Data (n = 985) from (Skjelkvåle et al. 1999).

Cadmium

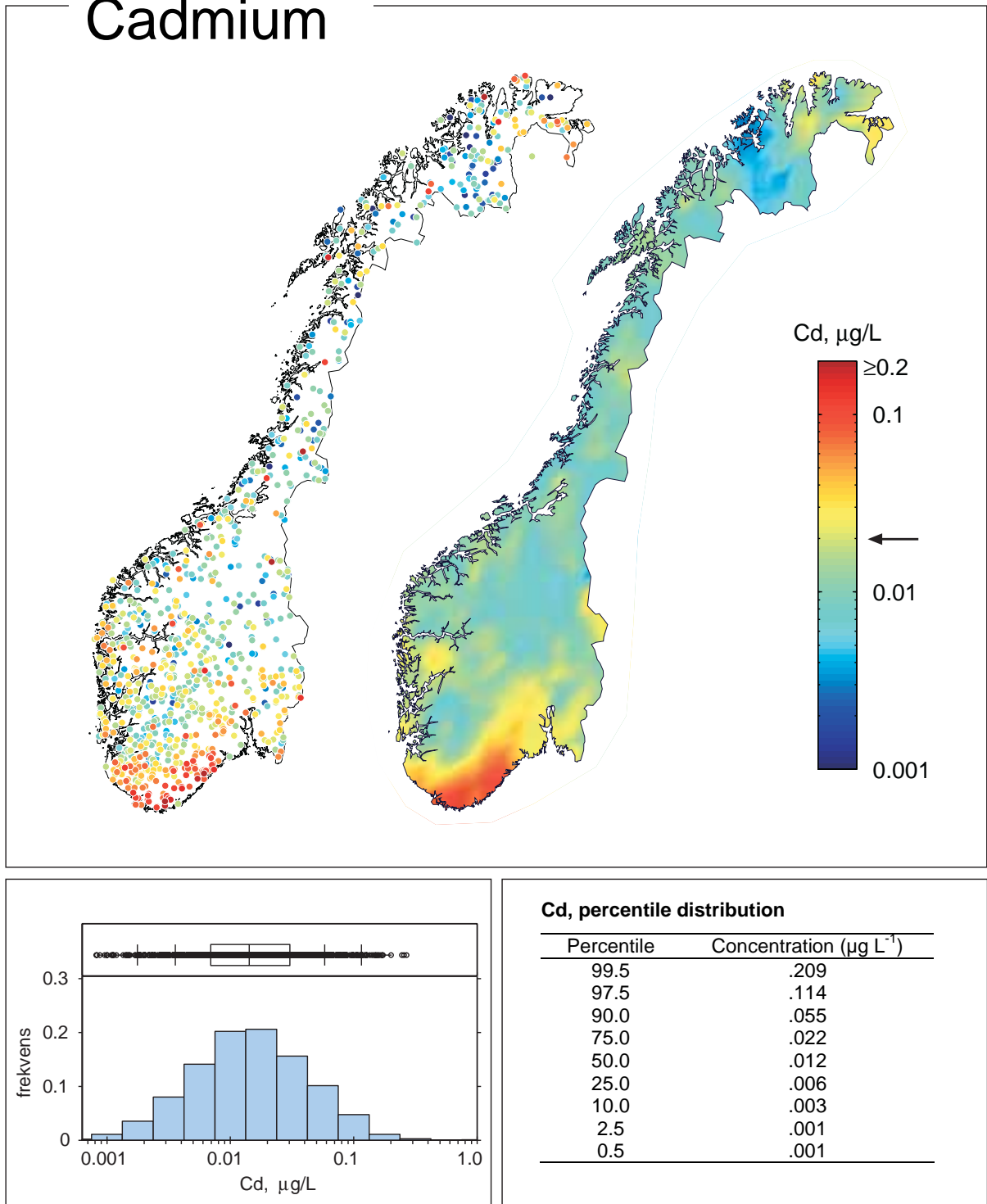


Figure 2. Concentrations of Cd in Norwegian lakes, autumn 1995. The map on the right side is based on spatial interpolation (kriging). Data (n = 985) from (Skjelkvåle et al. 1999). The arrow on the concentration scale indicates detection limit for the analytical method. Values below this point are estimated (see procedure in Skjelkvåle et al. 1999).

3. Critical limits

The concentrations of lead and cadmium in Norwegian lakes are in general below the suggested critical limits to be used in calculations of critical loads. The critical limits suggested in the manual for critical loads calculations are $0.3 \mu\text{g L}^{-1}$ for Cd and $11 \mu\text{g L}^{-1}$ for Pb.

At an ICP Waters workshop in March 2002 (Skjelkvåle and Ulstein 2002) available data on critical limits from a range of sources were summarized. It was concluded that the critical limits are in the range $0.1\text{-}1.0 \mu\text{g L}^{-1}$ for Cd and $1\text{-}10 \mu\text{g L}^{-1}$ for Pb (**Table 1**). The values suggested in the critical loads mapping manual are within these ranges, although that for Pb in the upper part of the range. Since Norwegian lakes typically have low concentrations of TOC and base cations, the critical limits for Norwegian lakes should most likely be in the lower part of the suggested range.

In this report we calculated critical loads using two different critical limits for each of the two metals. Both the limits suggested in the Mapping Manual and the lowest value in the range suggested at the ICP Waters workshop are used (**Table 1**).

Table 1. Recommended critical limits for Pb and Cd in surface water as recommended in the Mapping Manual (UBA 2003) and the ICP Waters workshop (Skjelkvåle and Ulstein 2002).

Metal	Critical limits from the Mapping Manual ($\mu\text{g L}^{-1}$)	Critical limits suggested at ICP Waters workshop ($\mu\text{g L}^{-1}$)
Cd	0.3 (0.2-0.4)	0.1-1.0
Pb	11 (9-13)	1-10

4. Model for critical loads calculations

The suggested steady-state model for the critical load of the metal M:

$$CL(M) = M_u - M_w + M_{ret(crit)} \cdot A_l/A_c + M_{lo(crit)}$$

M_u is net uptake: at present we set this to 0 assuming no biomass removal (i.e. no logging). This gives the most conservative estimate for the critical load. (unit: $g\ ha^{-1}\ a^{-1}$)

M_w is weathering. In the most recent manual (UBA 2003) this term is suggested ignored. In the previous version of the manual (de Vries et al. 1998) the weathering term is suggested calculated as a fraction of the base cation weathering (unit: $g\ ha^{-1}\ a^{-1}$). This presupposes that base cation weathering, as well as the content of heavy metals and base cations in the bedrock, is known and that these are proportional. This is not the case for Norway. In order to estimate the contribution of Pb and Cd in surface waters from weathering we assumed that in areas with low atmospheric deposition and no local sources, all the Pb and Cd in water comes from weathering. The weathering term is low and the assumption in the new manual, in which $M_w = 0$, seems reasonable.

$M_{ret(crit)}$ is the net retention of the metal in the lake (unit: $g\ ha^{-1}\ a^{-1}$). This term is suggested calculated from measured total concentration in water ($[M]_{tot,sw(crit)}$; unit: $mg\ m^{-3}$) and a net retention factor for the lake (r_{ret} ; unit: m/yr):

$$M_{ret(crit)} = 10 \cdot r_{ret} \cdot [M]_{tot,sw(crit)}$$

A_l/A_c is the lake to catchment area ratio.

$M_{lo(crit)}$ is the critical limit (flux) (unit: $g\ ha^{-1}\ a^{-1}$)

Two different approaches are suggested. One is an effects-based approach parallel to the method for acidification. Here critical limits for concentrations in waters are used. The other approach is called the stand-still approach and stipulates that the metal concentration in the future does not increase above the current level.

The manual takes into account the difference between total concentration and dissolved concentration, as it is assumed that the dissolved fraction only is responsible for effects on biota. In Norway only total concentrations are analysed. As Norwegian lakes typically are relatively clear with low concentrations of suspended matter, we take the total concentration as a measure of the dissolved concentration. In addition, comparing the total concentrations instead of dissolved concentrations with the critical limit will give a conservative estimate for the critical loads.

5. Selection of input data

We use the measured total concentrations of Pb and Cd from the national survey in 1995 (Skjelkvåle et al. 1999). In total 985 lakes were analysed for Pb and Cd in this survey. The data set also contains information on specific discharge, catchment and lake size and percent forest cover in each catchment.

Regarding in-lake retention, there are estimates of sedimentation velocities available for Norwegian lakes, but not from the lakes analysed in the 1995 survey. Annual sediment accumulation rates in sub-alpine lakes have been estimated to 0.5 ± 0.3 mm and in Nordic forest lakes 1.2 ± 0.5 mm (Rognerud and Fjeld 2001). As a starting point, we have used 0.5 mm for lakes with less than 20% forest in the catchment and 1.2 mm for lakes with more than 20% forest in the catchment.

Regarding weathering rates, we have estimated this from concentrations in assumed background areas. The maps indicate background concentrations about $0.1 \mu\text{g/l}$ for Pb and $0.01 \mu\text{g/l}$ for Cd. Alternatively the average concentrations for all lakes north of the Sognefjorden (61.5 degrees N) are $0.12 \mu\text{g/l}$ for Pb (omitting the sample from the high concentration lake in Mo i Rana) and $0.011 \mu\text{g/l}$ for Cd. For lakes having concentrations lower than these values, a weathering rate of 0 was used in the calculations.

6. Results

Critical loads calculated using two different critical limit and stand-still loads are presented for Pb in **Figure 3** and **Figure 6** and for Cd in **Figure 4** and **Figure 7**. The critical loads using the two different critical limits are compared in **Figure 5**.

There are large variations in the results for the three different approaches. The critical loads using the critical limits from the Mapping Manual give the highest critical load values, while the stand-still loads in general give very low values.

According to EMEP calculations the current deposition in southern Norway is about 10-15 g ha⁻¹ yr⁻¹ for Pb and 0.5-0.7 g ha⁻¹ yr⁻¹ for Cd (Berg et al. 2003¹). For both metals the deposition is in general lower than the critical load using the critical limit from the manual, while using the low critical limit the deposition is of similar size as the critical loads. The stand-still loads are in general lower than the deposition. For cadmium the stand still loads are 0 for the majority of the lakes.

For the effects based approach the choice of the critical limit becomes determinant for the critical loads (**Figure 5**). When changing the critical limit, the critical load is influenced by a factor of similar size. The points for Cd form two lines, dependent on whether or not the lake concentration was below the assumed concentration originating from weathering.

The clear geographical pattern seen on the maps for the effects-based approach reflects the gradient in the water discharge used in conversion from concentration to flux for the critical limit.

For the stand-still approach the maps show very low critical load values. This is because the present concentrations in general are low. Hence nearly no deposition is allowed without increasing the current concentrations. The pattern seen on the stand-still map for Pb reflects the pattern in the measured concentrations; if the current concentration is high, a higher deposition level can be tolerated without increasing the current concentrations.

In the most recent version of the draft Mapping Manual it is suggested not to include weathering except in special cases, and to use streams to avoid taking in-lake retention into account. The calculations presented here indicate that these two terms give minor contributions to the critical load values and hence can be omitted for simplicity in Norwegian lakes.

Heavy metals occur in a number of chemical species in water. For practical reasons these are usually defined in terms of analytical methods used. The total concentration is comprised of particulate and dissolved fractions. The dissolved fraction is in turn comprised of complexed or polymorphous forms and fractions. Generally the free ion fraction is the fraction of biological significance. The data from the Norwegian survey include only total concentrations.

There are efforts internationally to develop a method for estimating free ion concentrations and to use these instead of the total concentrations. Using the free ion concentrations would change the critical loads. The free ion approach should be tested on the same data as have been used here when available.

¹ See also www.msceast.org

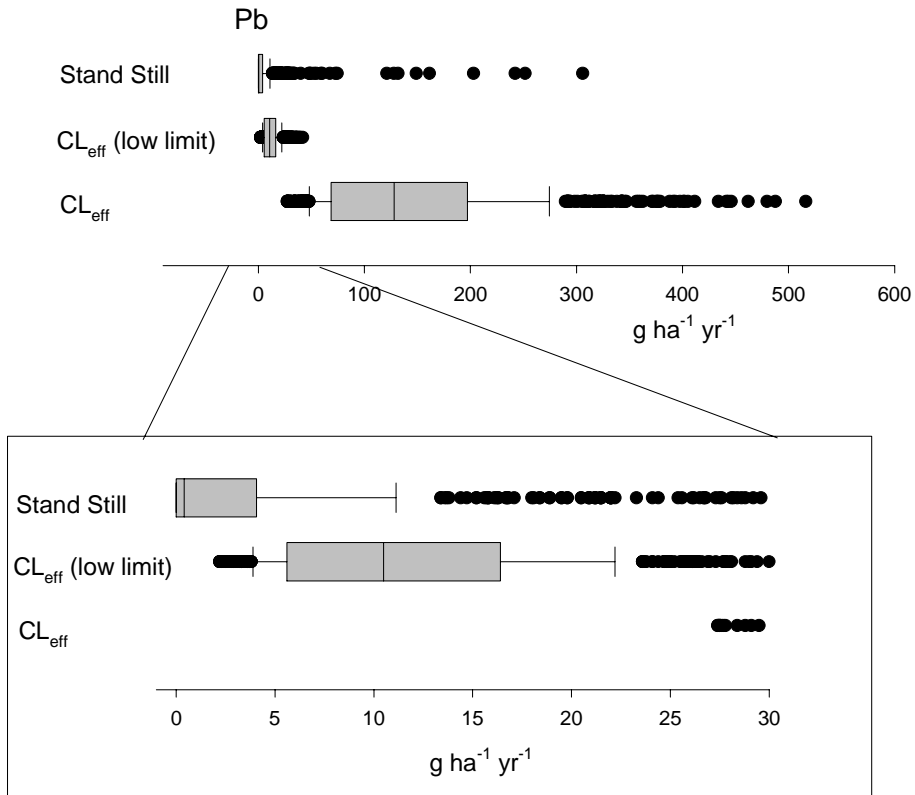


Figure 3. Distribution of values for critical loads for Pb calculated using the stand still approach and the effects based approach. The lower panel shows a segment for the lower part of the axis. For comparison: the annual deposition load in southern Norway is $10\text{-}15 \text{ g ha}^{-1}$.

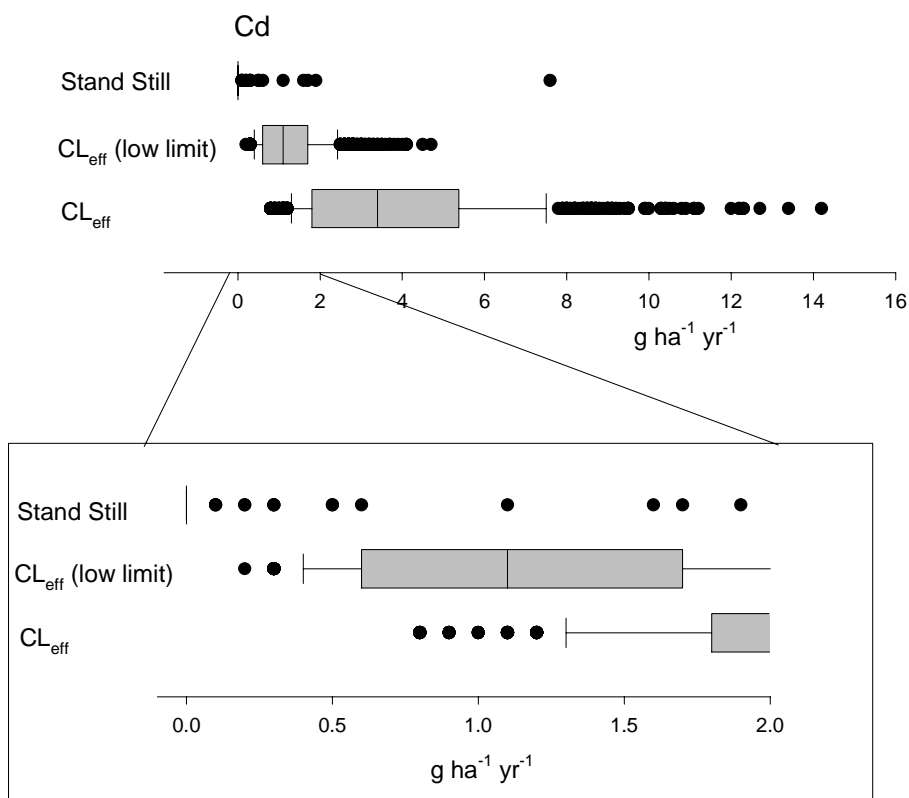


Figure 4. Distribution of values for critical loads for Cd calculated using the stand still approach and the effects based approach. The lower panel shows a segment for the lower part of the axis. For comparison: the annual deposition load in southern Norway is 0.5-0.7 g ha⁻¹.

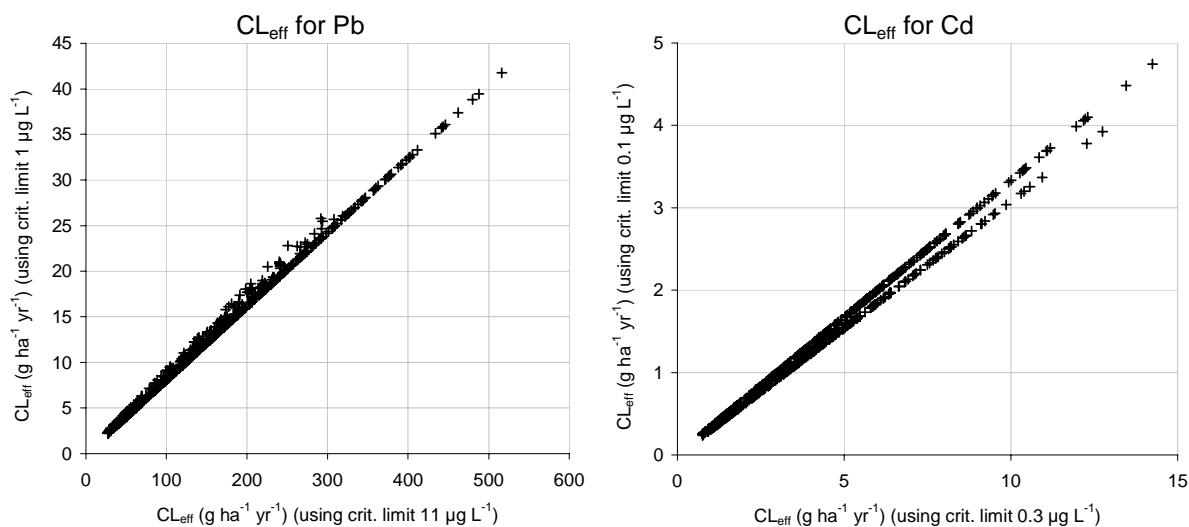


Figure 5. Comparison of critical loads for Pb and Cd calculated using two different values for the critical limits. The values on the x-axis are critical loads using the critical limit value from the manual; the y-axis the lower value of the critical limits range suggested by the ICP Waters workshop (Skjelkvåle and Ulstein 2002).

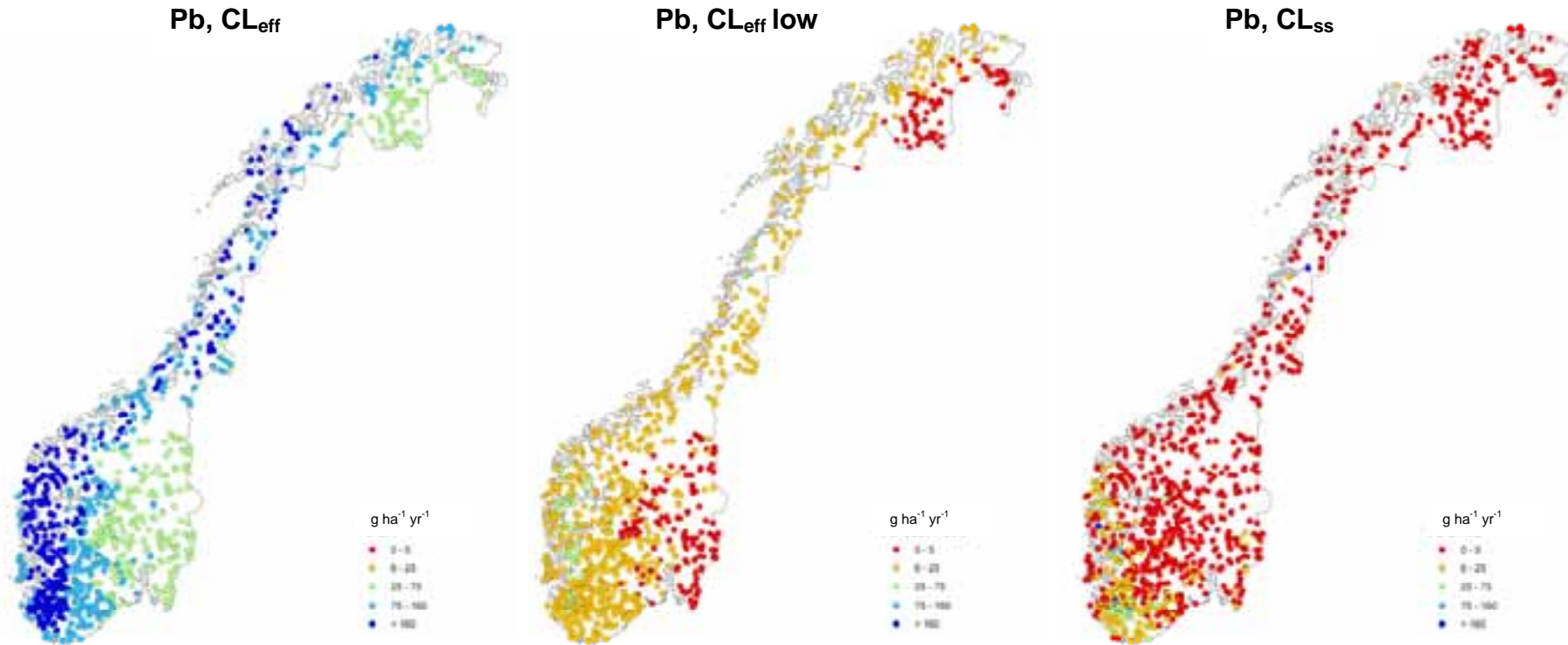


Figure 6. Critical loads and stand still loads for Pb. The left panel shows the critical loads using the critical limit suggested in the Mapping Manual (CL_{eff}), the middle panel the critical loads using the low critical limit ($\text{CL}_{\text{eff low}}$), and the right panel the stand-still loads (CL_{ss}).

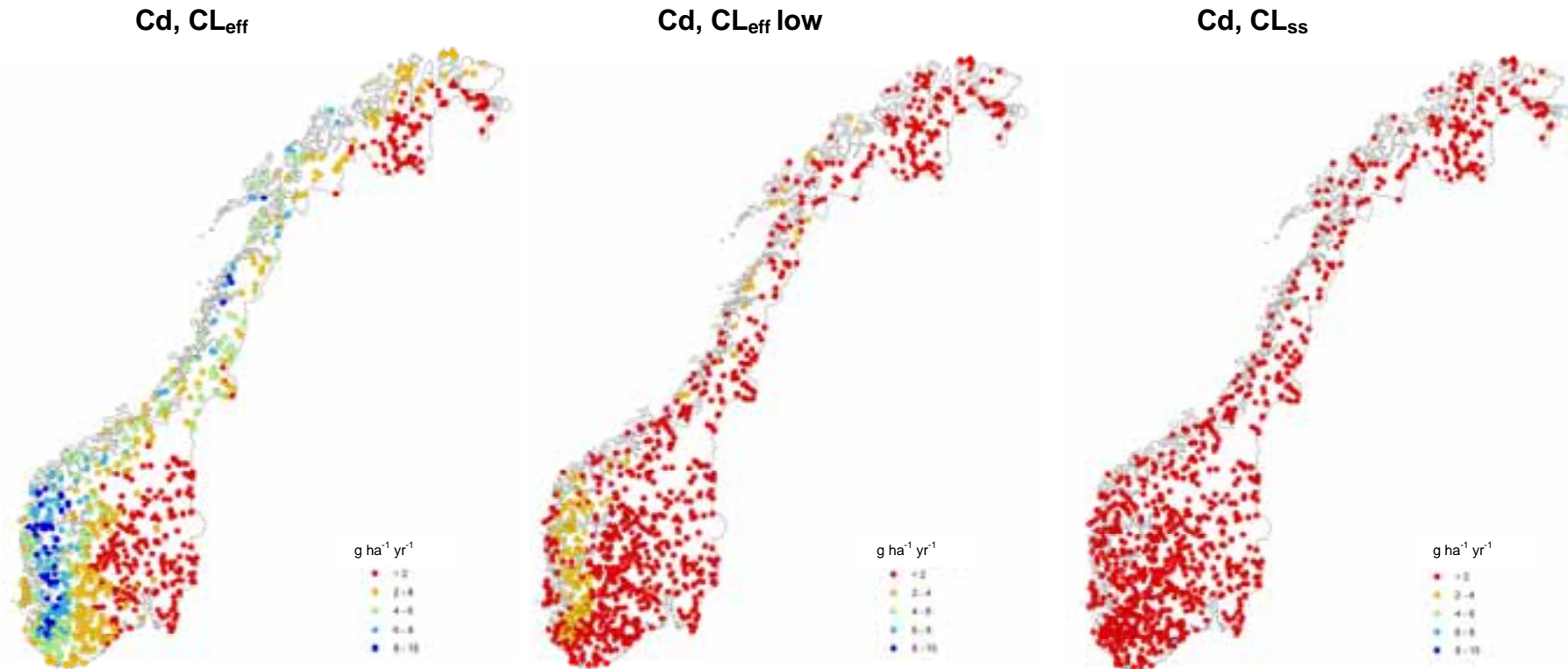


Figure 7. Critical loads and stand still loads for Cd. The left panel shows the critical loads using the critical limit suggested in the Mapping Manual (CL_{eff}), the middle panel the critical loads using the low critical limit ($\text{CL}_{\text{eff low}}$), and the right panel the stand-still loads (CL_{ss}).

7. Conclusions and recommendations

The measured total concentrations of Pb and Cd in 985 lakes are, with a few exceptions, lower than the recommended critical limits.

The critical loads for Pb and Cd are highly dependent on the values used for critical limits. Using the limit recommended in the draft Mapping Manual, the critical loads are high compared to the deposition. Using critical limits in the low range of suggested critical limits, the deposition are of similar size as the critical loads.

The stand-still approach permits very little deposition of both Pb and Cd. For Cd zero deposition is permitted most places, while for Pb deposition of a few $\text{g ha}^{-1} \text{yr}^{-1}$ is permitted, considerably less than the current deposition.

The methodology should be evaluated further, in particular, the free ion approach should be evaluated for Norwegian waters.

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Nature Tolerance Levels

The programme on Tolerance Levels in Nature was started by the Norwegian Ministry of Environment in 1989. The programme aims to obtain background material for international agreements on reductions of emissions. Within the Convention on Long Range Transboundary Air Pollution the members have decided that new agreements on emission reduction will be based on the principle of critical load.

A steering group with members from the Ministry of Environment has the overall responsibility of the programme.

The administration of the programme has been given to a working group with representatives from the Directorate for Nature Management (DN) and Norwegian Pollution Control Authority (SFT).

The working group has the following representatives:

Tor Johannessen - SFT
Else Løbersli - DN
Steinar Sandøy - DN

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