POTENTIAL EFFECTS OF METALS IN REACIDIFIED LIMED WATER BODIES IN NORWAY AND SWEDEN

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(Received 20 June, 2000; accepted 21 November, 2000)

Abstract. The goal of this work was to assess risk of chemical and biological effects of metals in reacidified, limed water bodies in Norway and Sweden. The risk assessment is based on a literature review and evaluations of water chemical data from the 1995 Nordic Lake Survey. Compared to the pre-liming period, it us unlikely that enhanced remobilization of inorganic aluminium (Al) or other toxic metals (metal bomb hypothesis) from the catchment, the lake sediment and/or the streambed will occur when limed waters reacidify. Rather, the concentrations in surface waters are expected to be lower than before liming started, because of reduced atmospheric inputs of both strong acids and metals as Cd, Hg, Pb, and Zn during the last 10–20 yr. The concentrations in lakes relative to the biological effect levels, as well as the chemical properties of the different metals suggest that the potential biological risks associated to reacidification of limed lakes decrease in the order Al >> Cd > Pb. The risks associated with Cr, Cu, Fe, Mn, Ni and Zn are very low and do not have to be considered except in waters with known concentrations larger than the lowest biological risk level. Such waters are very rare (<2%). Aluminium is the metal that should be used to set the limit for judging the risk of biological damage due to reacidification of limed surface waters.

Keywords: acid lakes, heavy metals, limed lakes, reacidification, Scandinavia

1. Introduction

Base addition, primarily lime, has been widely practised to mitigate surface water acidification in Sweden, Norway, Canada and the U.S.A. Besides increasing pH and the acid neutralising capacity of water (ANC), liming also reduces the concentration of toxic Al so that surface water ecosystems again are able to host viable fish populations. It is unclear to what extent liming also has reduced the concentration of other potentially toxic metal-species. Due to generally low concentrations, however, effects of liming on other metals have so far been less recognised. This is reasonable, because metals other than Al only have caused local damages directly related to point sources of those metals.

In Sweden, liming has received governmental subsidies since 1976, and by 1999, 2 billion SEK has been used on different liming activities. During last five years, approximately 200 000 tons of lime have been used in lakes (n = 7500), rivers (12 000 km) and on wetlands (n = 1600 objects), corresponding to approx-

Environmental Monitoring and Assessment **73:** 155–178, 2002.

imately 175 million SEK per year. In Norway, about 2000 localities, ranging from small lakes to large salmon rivers have been limed, but only a few experimental wetland and catchment liming projects have been conducted.

Recently, the Swedish liming programme has been evaluated, and a large financial reduction down to 80 million SEK a year was proposed (SOU 1996:53). The budget never reached such low levels, and for 1999 the budget was reduced to 150 million SEK. Since 1998, there has also been a cut in the Norwegian liming program, from 119 million NOK in 1997, to 105 million NOK in 1998. Accordingly, concerns have been raised on the chemical and biological consequences of reacidification of previously limed systems.

The goal of our work was to make a risk assessment of the chemical and biological effects of metals in reacidified, limed water bodies. Most of the water chemical data used in our work are from the 1995 Nordic Lake Survey (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999). We deal with the most relevant metals (Al, Cd and Pb) with respect to biological effects in Norwegian and Swedish surface water.

2. Trends in Atmospheric Deposition

In Norway, the mean concentrations of sulphate (SO_4^{2-}) in precipitation have decreased by 39–58% at 11 precipitation stations during the period 1980–1996 (Tørseth and Manø, 1997). During the same period no significant changes have been recorded for nitrate (NO_3^-). For ammonia (NH_4^+) 3 stations show decreasing trends, one increasing, while the remaining stations show no significant changes. At 25 stations distributed all over Sweden the reduction in SO_4^{2-} has been approximately 50%, with relatively small differences in trends between the stations, during the period 1983–1995. The sulphate deposition was at its maximum in the early 1970s (L. Granath, MISU, pers. com.). A future deposition scenario for the next decade indicates that S-deposition will level out at slightly lower level than at present in Norway and Sweden (Mylona, 1996; Berge, 1997).

Norway has weekly wet-deposition data of heavy metals from 3 weather stations for Cd, Pb and Zn back to 1978. These data show a 60–80% concentration decrease of Pb in wet-deposition from 1978 to 1996, and a decrease of 70% for Zn and 50–80% for Cd (Tørseth and Manø, 1997). The concentrations of the same metals in wet-deposition have also decreased significantly at four precipitation stations in Sweden during 1984–1995, i.e. by 25–53% and 48–76% for Cd and Pb, respectively. For Zn, significant downward trends (23–35%) were only documented at Arup and Svartedalen, in southwestern Sweden. No significant trends could be detected for Cu at any of the Swedish stations (Lydersen and Löfgren, 2000).

The reductions in atmospheric inputs of both strong acids and heavy metals during the last couple of decades have also caused significant improvements of surface water chemistry (Table I). In 70% of the Swedish reference lakes (188

TABLE I

Mean values of key water chemical parameters in 485 lakes in Norway (N) sampled both in 1986 and 1995

Area	SO_4^*		ANC		$(CM)^*$		pH		LAL	
	1986	1995	1986	1995	1986	1995	1986	1995	1986	1995
	(μeq L^{-1})						(–log[H^+])		(μg L^{-1})	
Eastern Norway	73	50	19	39	79	78	5.44	5.65	42	28
Southern Norway	60	36	–28	–12	29	22	4.82	4.97	112	64
Western Norway	30	23	–8	1	26	21	5.24	5.39	22	15
Mid Norway	16	12	17	26	32	30	6.09	6.15	4	1
Northern Norway	47	42	24.5	43	73	75	6.45	6.52	0	0

SO_4^*: non-marine SO_4^{2-}; ANC: acid neutralising capacity; $(CM)^*$: non-marine Ca^{2+} and Mg^{2+}; LAL: labile aluminium.
Table modified from Skjelkvåle *et al.* (1996).

lakes), the alkalinity has increased with a median for all reference lakes of 1.4 μeq L^{-1} between 1983 and 1994 (Wilander, 1997). By 1997 there was a positive trend for alkalinity in 87% of the reference lakes (Wilander, unpubl. data). A comparison between the 1990 and 1995 lake surveys also shows increasing alkalinity in unlimed lakes (Wilander *et al.*, 1998). These improvements have to be considered in the risk assessment and in light of the future water chemistry in waters that may reacidify due to terminated liming.

3. Liming Strategies and Chemical Effects

The liming criteria in Norway and Sweden are different. In Norway negative biological effects have to be documented, and a target pH of 6.0 is defined as a proper liming goal, except in salmon rivers during the smoltification period (late winter and spring), when somewhat higher pH (up to 6.4 to 6.5) is recommended. Lakes with acid neutralising capacity (ANC) > 20 μeq L^{-1} ($ANC = [\Sigma Ca^{2+}, Mg^{2+}, Na^+, K^+] - [\Sigma SO_4^{2-}, NO_3^-, Cl^-]$) are normally not limed in Norway, as well as humic lakes with high concentration of organic acids, i.e. total organic concentration (TOC) > 6–8 mg C L^{-1}, regardless of pH.

In Sweden, lakes with pH < 6.0 and/or inorganic alkalinity <50 μeq L^{-1} are candidates for liming, and alkalinity $\geq$100 μeq L^{-1} is the chemical goal for liming. Negative biological effects do not have to be documented. Average alkalinity of 634 limed lakes was 50 μeq L^{-1} prior to liming and the lowest alkalinity of these lakes was 30 μeq L^{-1} (Table II). The high colour (average: 66 mg Pt L^{-1}) in many of the lakes indicates that ANC would have been much higher than the alkalinity.

TABLE II

Average values of pH, alkalinity, the sum of Ca and Mg, and water colour before and after liming in 634 lakes in Sweden. Colour is divided into lakes with colour <100 mg Pt L^{-1} before liming and >200 mg Pt L^{-1} before liming. Table modified from Thörnelóf and Degerman, 1991

Parameter	Before liming	After liming	*t*-test
Average pH	5.83	6.42	$p < 0.001$
Lowest pH	5.67	6.20	$p < 0.001$
Average alkalinity (meq L^{-1})	0.05	0.15	$p < 0.001$
Lowest alkalinity (meq L^{-1})	0.03	0.11	$p < 0.001$
Average conductivity mS m^{-1}	4.90	5.20	$p < 0.001$
Average Ca^{2+} + Mg^{2+} (meq L^{-1})	0.27	0.35	$p < 0.001$
Average colour (mg Pt L^{-1})	66.6	74.5	$p < 0.001$
Colour when pre-treatment colour <100 mg Pt L^{-1}	46.0	62	$p < 0.001$
Colour when pre-treatment colour >200 mg Pt L^{-1}	246	206	$p < 0.001$

Direct liming of surface waters causes a substantial and rapid change of the water chemical conditions, primarily due to increased pH, alkalinity, ANC and Ca^{2+}. The subsequent effect of liming is reduction in the concentration of inorganic aluminium forms (LAL), the primary toxic compounds in acid surface waters. In some cases reductions in other potentially toxic metals (e.g. Cd, Fe and Mn) occur after liming (Svensson *et al.*, 1995; Wilander *et al.*, 1995). Direct lake liming does not prevent episodic acidification of stream water and the near-shore region of the lake during snowmelt, a phenomenon critical for the development of fish population. Furthermore direct surface water liming does not reduce the inputs of toxic Al-forms from groundwater or upstream.

Watershed liming ($CaCO_3$) results in a gradual increases in pH, alkalinity (or ANC) and Ca^{2+} in the water column (Driscoll *et al.*, 1996; Hindar *et al.*, 1996; Abrahamsen, 1995). Watershed liming eliminates the episodic acidification of stream water and the near-shore region of the lake during snowmelt. Watershed liming generally decreases the transport of Al to lake and running waters, and increases the concentrations of dissolved organic carbon (DOC) and dissolved silica (H_4SiO_4) in stream and lake waters.

4. Metal Concentrations in Relation to Biological Effects

The concentration of metals in Swedish and Norwegian lakes are generally *very low* to *low* according to the freshwater classification systems in Sweden and Norway (SFT, 1997; Alm *et al.*, 1998). Very few lakes have metal concentration above or close to concentrations where biological effects are expected (lowest biological

TABLE III

Concentrations (percentiles and lowest biological risk levels in μg L^{-1}) of heavy metals and arsenic in Swedish and Norwegian non-limed lakes in 1995. The lowest biological risks levels (LBRL) used are based on the lowest assumed effect criterion in Norway (SFT, 1997) and Sweden (Alm *et al.*, 1998). Data are from Henriksen *et al.* (1998) and Skjelkvåle *et al.* (1999)

	Pb	Cd	Cu	Zn	Ni	Cr	Fe	Mn	Co	As[a]
Maximum	14.97	1.072	37.70	426.5	10.17	27.68	10149	697.4	3.151	5.054
90%	0.82	0.057	1.04	6.34	0.92	0.38	557.9	51.4	0.203	0.373
75%	0.39	0.028	0.62	3.11	0.53	0.20	221.5	17.9	0.097	0.174
50%	0.16	0.012	0.37	1.43	0.29	0.08	71.2	5.4	0.049	<0.1
25%	0.08	0.006	0.22	0.69	0.15	0.04	23.8	1.8	0.023	<0.1
10%	0.04	0.003	0.14	0.29	0.08	0.02	9.8	0.7	0.010	<0.1
Minimum	0.00	0.000	0.02	0.07	0.01	0.00	0.1	0.0	0.001	<0.1
LBRL	>1	>0.1	>3	>20	>15	>5	no	no	no	>5.0

[a] Sweden only.

risk levels = LBRL). Only LAL in surface waters is a regional problem for aquatic life in both countries (Table III).

Regarding LAL, no official LRBL is established. However, Polèo *et al.* (1997) reported relative sensitivity among common Scandinavian fish species to acute Al challenges to be in the order:

Atlantic salmon (*Salmo salar*) > roach (*Rutilus rutilus*) > minnow (*Phoxinus phoxinus*) > perch *(Perca fluviatilis*) > greyling (*Thymallus thymallus*) > brown trout (*Salmo trutta*) > arctic char (*Salvelinus alpinus*) » crucian carp (*Carassius carassius*).

Although there is a considerable range in the sensitivity to Al among fish species, salmonids are the most sensitive group, particularly at the early gill-breathing stages in their life (Schofield, 1977; Baker and Schofield, 1982; Cleveland *et al.*, 1986). The embryo is the life stage least sensitive to Al (Leivestad *et al.*, 1987), while the smoltification period of Atlantic salmon likely is the most sensitive stage among all Scandinavian fish species (Staurnes *et al.*, 1993). There are large strain differences towards Al/acidification (Dalziel *et al.*, 1995), and the strains that naturally have survived in the most acidified areas of Norway seem to be the most tolerant. This indicates adaptation as an important mechanism.

Neither LAL, TOC nor pH are the best single factor explaining fish status in Norwegian surface waters (Bulger *et al.*, 1993; Lien *et al.*, 1996; Lydersen *et al.*, 1994b). The best variable is ANC as defined by Reuss and Johnson (1986). The Norwegian water chemistry and fish status studies (including brown trout, arctic char and perch) are based on approximately 1000 Norwegian lakes sampled at autumn turnover. These studies concluded that for all lakes with ANC $\geq$ 20 μeq

L^{-1}, damage to fish populations of the three species studied is rare. Nominally, this means that all lakes with ANC $\geq$ 20 μeq L^{-1} during autumn turnover always have sufficient acid neutralising capacity to avoid episodes affecting these fish species at population level. The invertebrate survey by Lien *et al.* (1996) concluded that ANC = 20 μeq L^{-1} is also a concentrations where no severe damage to invertebrate fauna has been recorded in Norwegian surface waters.

Lydersen *et al.* (1994b) found that at pH $\leq$ 4.75, the LAL concentration is almost always > 50 μg Al L^{-1} and most lakes have extinct or reduced fish populations. However, the ionic strength (often expressed with Ca-concentration) is important for the toxicity, a well-known phenomenon (Cleveland *et al.*, 1986; Wood *et al.*, 1988, 1990). Lakes with pH $\geq$ 6.0±0.25 and Ca-concentrations $\geq$ 2 mg Ca L^{-1}, seldom have damaged fish populations of brown trout, arctic char and perch, and extinction is unlikely (Lydersen *et al.*, 1994b). Only some of these lakes have LAL-concentration > 80–100 μg Al L^{-1}. Based on the total population of non-limed lakes analysed for pH and Ca in Sweden during the 1995 survey (n = 3302), only 15% of the lakes had Ca < 2 mg L^{-1}, and only 7.2% of the lakes had pH < 5.75.

TOC also plays an important role for fish status. It is rather uncommon that fish population are extinct or reduced in Norwegian lakes with high TOC and pH $\leq$ 6±0.25 (Lydersen *et al.*, 1994b). Besides higher LAL at pH < 5, there is also a tendency of higher LAL with higher TOC within the pH-interval 5–6 (Figure 1).

There may be a critical concentration of inorganic Al (LAL) within the range 20–80 μg Al L^{-1}, where sublethal effects occur. Effects of sublethal Al-concentration are reduced hatchability, reduced larval feeding behaviour, swimming capacity, slower growth, and general poorer condition.

The most sensitive aquatic fish-species is Atlantic salmon (*Salmo salar*), with reported effects at about 20 μg Al L^{-1} (Staurnes *et al.*, 1995). Since roach (*Rutilus rutilus*) seem to be the second most sensitive fish specie in Nordic waters (Poléo *et al.*, 1997), we assume the same risk level for this specie. However, for waterbodies that never have hosted these species, somewhat higher LAL-concentrations should be acceptable. Even though we are aware of tolerance differences between species and strains, we assume LAL-concentrations > 80 μg Al L^{-1} (Lydersen *et al.*, 1994b) to be highly critical for freshwater fishes as minnow, perch, grayling, brown trout, and arctic char. Thus, effects on fish species might be expected within the LAL-concentration range: (20–80 μg Al L^{-1}). Accordingly, many lakes in Norway and Sweden (Table IV) have LAL concentrations above the lowest biological risk level (LBRL) for LAL. Most of these lakes have low pH and/or medium to high TOC.

Table V shows the percentage of non-limed lakes in Norway, Sweden and Finland, and limed Swedish lakes *below* LBRL for different metals. Excluding Cd, Pb and Al, <2% of the lakes have heavy metal concentrations > LBRL. For Cd, Pb and Al, 5, 7 and 19% of the non-limed Swedish lakes have concentrations > LBRL (according to the Swedish LBRL-criteria for Cd and Pb). The differences

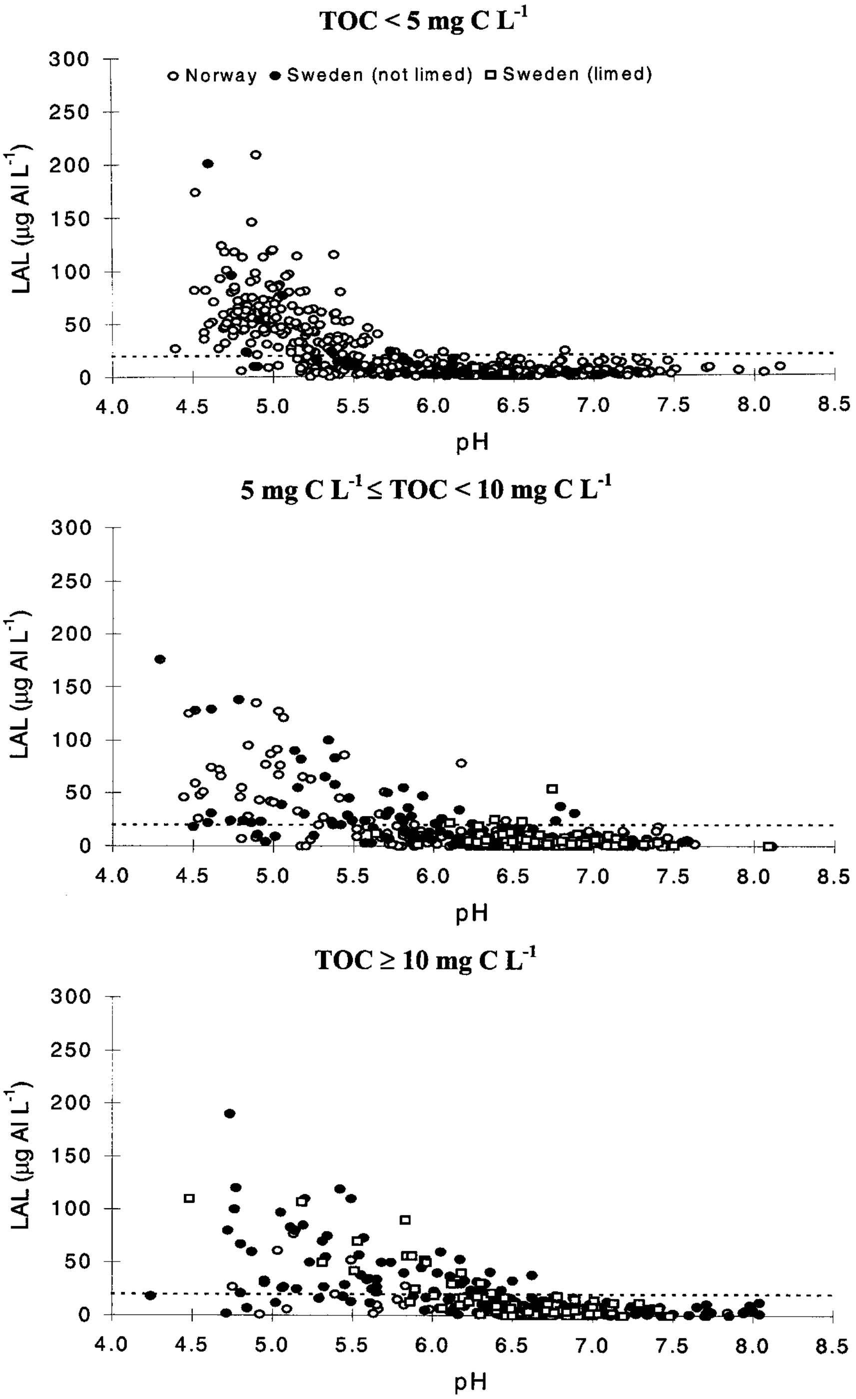

Figure 1. Concentrations of labile aluminium (LAL) in Norwegian and Swedish non-limed and limed lakes in relation to pH for 3 different TOC-classes. The dotted line indicates reported effects on aquatic organisms, LAL = 20 μg Al L^{-1}. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

TABLE IV

Percentile distribution of dissolved inorganic Al (LAL) in Norwegian and Swedish surface waters, based on the Nordic Lake survey in 1995. Norway: n = 969; Sweden, non-limed: n = 555; Sweden, limed: n = 157. Values < 10 μg Al L^{-1} are below the detection limit for the Norwegian method. (Suggested lowest biological risk level (LBRL) of 20 μg LAL L^{-1} is related to Atlantic salmon, based on the work by Staurnes *et al.*, 1995). Data are from Henriksen *et al.* (1998) and Skjelkvåle *et al.* (1999)

Percentile	LAL		
	Norway Non-limed	Sweden Non-limed	Sweden Limed
	(μg Al L^{-1})		
Maximum	174	201	110
99.5%	125	149	110
97.5%	89	90	58
90.0%	53	34	24
75.0%	10	14	11
50.0%	5	5	5
25.0%	5	0	0
10.0%	2	0	0
2.5%	0	0	0
0.5%	0	0	0
Minimum	0	0	0

are small between limed and non limed lakes, even though liming seemed to decrease the exceedance to 2 and 14% of the lakes for Cd and Al, respectively, while the percentage of lakes > LBRL are almost equal for Pb in limed and non-limed Swedish lakes. In Norway the percentage amounts of non-limed lakes above the same LBRL-values are somewhat higher for Al and Pb compared with the Swedish lakes, while somewhat lower for Cd.

The population of limed Swedish lakes generally has somewhat higher concentrations of heavy metals than the non-limed lakes. This may indicate that many of the most acidified and metal rich lakes already are limed, but also that liming has marginal effect on the total concentrations of heavy metals. Liming primarily change the physical-chemical forms of many metals, but this is of course important since toxicity is highly related to species of metals, and liming often reduce the most toxic metal forms. The limited number of reacidified limed lakes do not show elevated concentrations compared to levels expected from the ambient pH level. We have no pre-limed metal data on these lakes, but it indicates that no

TABLE V

Percentage of non-limed lakes in Norway (n = 985; n = 514 for As), Sweden (n = 820, n = 1124 for As) and Finland (n = 462), and limed Swedish lakes (n = 261) *below* the lowest biological risk levels (LBRL) according to Norwegian (SFT, 1997) and Swedish criteria (Alm *et al.*, 1998). The concentration for Al is labile aluminium (LAL), the toxic fraction of Al. No LBRL-values for LAL are established in Norway or Sweden. Thus, the tentative LBRL-values for Atlantic salmon (20 μg Al L^{-1}) and brown trout (80 μg Al L^{-1}) are according to the studies by Staurnes *et al.* (1995) and Lydersen *et al.* (1994b)

Element	Country	LBRL	Norway Non-limed	Sweden Non-limed	Sweden Limed	Finland Non-limed
		(μg L^{-1})		(%)		
Zn	Norway	50	99.8	99.6	99.6	99.8
	Sweden	20	99.3	99.3	98.9	99.4
Cd	Norway	0.2	99.5	98.6	98.9	99.8
	Sweden	0.1	96.6	95.5	97.7	99.4
Cu	Norway	3.0	99.3	98.4	99.6	99.1
	Sweden	3.0	99.3	98.4	99.6	99.1
Pb	Norway	2.5	98.4	98.2	98.1	99.8
	Sweden	1.0	91.5	93.0	93.5	98.3
Ni	Norway	5	99.8	99.6	99.6	98.9
	Sweden	15	100	99.8	100	99.4
Cr	Norway	10	100	99.8	100	100
	Sweden	5	99.9	99.6	100	100
As	Norway					
	Sweden	5	99.8	99.8	100	100
Al	*Atlantic salmon*	*20*	*76.9*	*80.7*	*86.0*	*n.a.*
	Brown trout	*80*	*95.5*	*91.4*	*92.4*	*n.a.*

severe increase in metal concentration should occur in many lakes when liming terminates. In addition, the concentrations of heavy metals are generally far from the lowest effect levels.

4.1. RELATIONSHIPS BETWEEN METAL CONCENTRATIONS, pH AND TOC

A redundancy analysis (RDA-analysis, i.e. a linear, multivariate method for describing more than one dependent variable in relation to several independent variables; Ter Braak, 1994) has been applied to evaluate trace metal concentrations in Swedish and Norwegian lakes against TOC and H$^+$. Lake chemistry data used are from Skjelkvåle *et al.* (1999). The RDA biplot (Figure 2) shows that the two independent variables are fairly orthogonal, and that they align closely with the

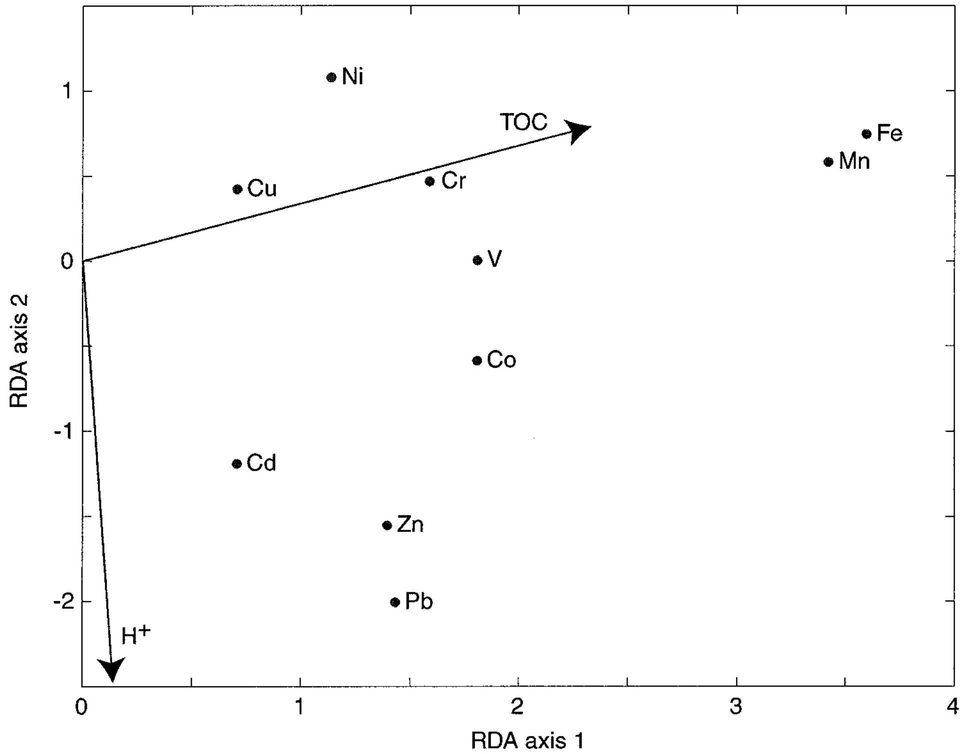

Figure 2. Redundancy analysis of trace metal concentrations against TOC and H^+ (arrows) for Swedish and Norwegian lakes based on the 1995 surveys. All variables are log-transformed. Trace metal concentrations below the detection limit are estimated according to Skjelkvåle *et al.* (1999).

RDA axes. The first RDA axis reflects a strong positive relationship between TOC and all the metals included in the analysis, but particularly Fe and Mn. The second RDA axis indicates a strong positive association between H^+ and Cd, Zn, and Pb, and perhaps a negative association with Ni. The strong association between H^+ and Pb is somewhat unexpected, due to much documentation in literature of strong associations between Pb and organic compounds. A problem with Cd, Pb and Zn is the important atmospheric inputs of these metals and the close relationship between atmospheric inputs of strong acids and these metals. This is likely the causal factor for the relationship between these metals and pH.

Because the RDA-analysis indicates that Cd, Zn and Pb are more pH sensitive, they are likely the most important metals with respect to reacidification effects. Inorganic Al is not included in the RDA-analysis but should be added to this list, since it is the most important element in acidic surface waters.

Concentration levels, pH dependency, and toxicity, all point to Pb, Cd and Al as the main metals to be concerned about when dealing with reacidification of limed lakes.

4.2. REMOBILIZATION OF METALS FROM SEDIMENTS

A literature review by Nelson and Campbell (1991) concluded that although laboratory experiments suggest that some trace metals could be remobilized from lake sediments at acidic pH, field studies indicate that sediment pore waters are not sufficiently acidic for metal remobilization to occur. Furthermore, pH in sediments is probably higher than the water column pH due to the internal alkalinity production in the sediments. Studies of epilimnetic sediments from 8 lakes (pH range 4.2–7.0) in Canada showed that pore water pH increased by sediment depth in the 6 lakes with a $\mathrm{pH} < 6.5$. The alkalinity (Gran titration in the pH range 4.2 to 3.5) in the sediments increased by greater than one order of magnitude above the water column values. The alkalinity increase started within 2 cm of the sediment-water interface and corresponded to an alkalinity flux out of the sediment of 0.4–510 meq m^{-2} yr^{-1} in the lakes with a $\mathrm{pH} < 6$ in the bottom water (Schiff and Andersson, 1986). Alkalinity might also be produced by sulphate reduction in sediments, increasing the pH and reducing the metal concentrations (Belzile and Morris, 1995).

The concentrations of LAL in the reacidified limed lakes included in the 1995 Swedish lake survey, indicate no dramatic reacidification effects, other than what is expected due to the reacidified pH-level. Similarly Hindar and Lydersen (1995) concluded that reacidification of limed surface waters, will not contribute to any dramatic increase in the concentration of Al. The concentration of inorganic Al will relatively rapidly return to pre-liming levels or lower. LAL-concentration lower than prelimed concentrations is likely since the amount of acidifying compounds in precipitation has decreased significantly during the last years. This has caused a significant reduction in LAL in most acidified surface waters in Norway (Table I). Accordingly, this is probably the case also in areas of Sweden where deposition of strong acids is the main cause of surface water acidification.

Lead sediment profiles from acidic lakes typically indicate that acidification has little effect on Pb accumulation in the sediments (Norton *et al.*, 1981; Nelson and Campbell, 1991). This has also been confirmed by laboratory experiments on surface sediments, where Pb release from sediments did not occur above pH 3 (Davis *et al.*, 1982). Even the transport of Pb within the lake and lake watershed is relatively unaffected by acidification. Regarding remobilization from Cd from sediments during reacidification, LaZerte (1986) concluded that after Cd is deposited, it is not readily released from aerobic sediments, except at very low pH ($\mathrm{pH} < 4.0$).

4.3. ALUMINIUM

Acidification of soilwater systems often causes increased transfer of Al from edaphic to aquatic environments (Cronan and Schofield, 1979; Dickson, 1980, 1983; Seip *et al.*, 1989). In the aquatic environment Al may be present in different physicochemical forms, varying for instance in size and charge properties (Hem and Roberson, 1967, 1990; Smith and Hem, 1972; Hem *et al.*, 1973; Lind and Hem, 1975; Schecher and Driscoll, 1987, 1988; Lydersen *et al.*, 1994a). The Al present in

TABLE VI

Percentage of non-limed lakes in Norway and Sweden (n = 1524) below the lowest biological risks levels (LBRL) of labile aluminium for Atlantic salmon (20 μg LAL L^{-1}) and for brown trout (80 μg LAL L^{-1})

pH interval	n	Atlantic salmon	Brown trout
pH: <5.4	242	38.0%	47.1%
pH: ≥5.4–6.0	199	89.9%	93.5%
pH: ≥6.0	1983	90.7%	96.4%

ionic form in natural water can normally be expected to occur as the free aquo Al^{3+}, hydroxide-, fluoride- and silicate-complexes, and as sulphate complexes in waters with low pH and high SO_4^{2-} concentrations. In waters of pH > 6, or in high organic carbon waters, the concentrations of these monomers are low, while polymerised Al-hydroxides and Al-organic aggregates of colloidal and subcolloidal size predominate.

The concentration and chemical forms of Al in soil and surface waters are highly dependent on pH, temperature and concentrations of inorganic and organic ligands. This is of major importance because the acute toxic forms of Al are the inorganic monomeric and polymeric Al-cations (Lydersen, 1992). This is reasonable since the gill surface seems to be the main target organ. Cation exchange success on the gill surface of aquatic organisms is decisive for Al-toxicity. A main reason why increased base cation concentrations reduce Al-toxicity is because they are non-toxic and present in much higher concentrations compared to the toxic metals. That means that both monovalent and divalent base cations compete well with Al (or other toxic cations) on active negative sites on gills, and accordingly reduce the metal toxicity.

Data from the 1995 Nordic Lake Survey shows that 23% of the Norwegian lakes and 19% of the non-limed Swedish lakes have LAL-concentrations higher than the expected lowest LBRL-value, i.e. 20 μg LAL L^{-1} (Tables IV and V). For lakes with pH > 5.4, still approximately 10% of the lakes have LAL concentration > 20 μg L^{-1}. In lakes with pH < 5.4, almost two-thirds (62%) of the lakes exceed the LBRL-value (Table VI and Figure 5). This confirms the importance of Al as the most important metal causing toxicity in acidified Norwegian and Swedish surface waters.

For limed Swedish lakes in the 1995 Lake Survey, about 15% of the lakes have LAL > 20 μg LAL L^{-1} (Figure 3), indicating that these limed lakes have not achieved optimal water chemical conditions for Atlantic salmon and/or roach. The reason why some limed lakes have LAL-concentrations > LBRL-value might be medium to high TOC concentrations, slow precipitation kinetics of LAL and/or

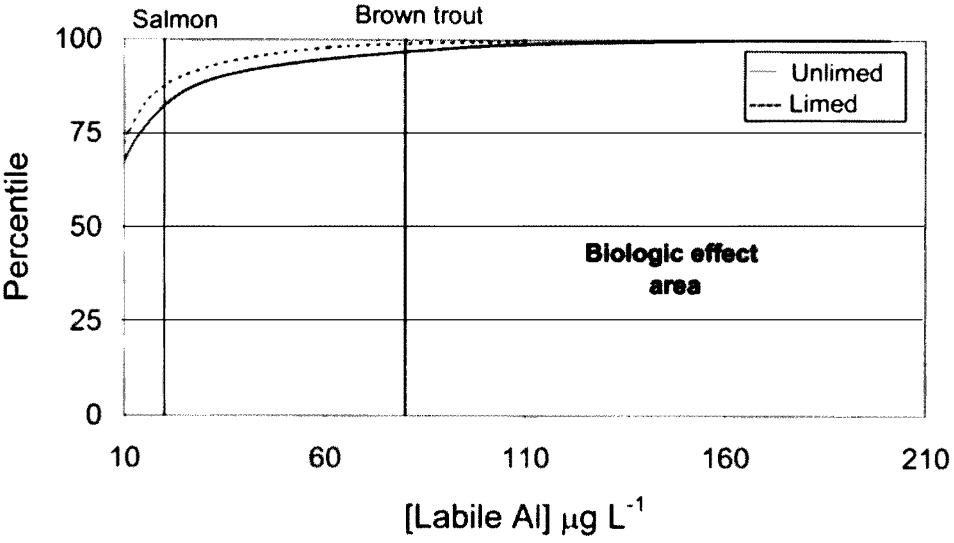

Figure 3. Percentile distribution of labile Al (LAL) in limed (n = 157) and not limed (n = 555) Swedish lakes. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

simply the reduced effect of liming by time in waters that are not continuously limed.

About 15–20% of the low TOC lakes (<5 mg C L^{-1}) and high TOC-lakes ($\geq$ 10 mg C L^{-1}) have LAL concentrations >20 μg Al L^{-1}, while about 35% of the medium TOC-lakes ($\geq$ 5–10 mg C L^{-1}) fell within this category (Figure 4).

The concentrations of LAL in reacidified limed Swedish waters incorporated in the 1995 Nordic Lake Survey indicate no dramatic effects of reacidification other than the expected. Thus, enhanced remobilization of LAL from the catchment, the lake sediment and/or the streambed should not be expected when limed objects reacidify, consistent with Hindar and Lydersen (1995). The concentration of inorganic Al will relatively fast be as before liming or even lower. There are at least two reasons why lower LAL-values are expected after reacidification:

(1) During a certain time period, reacidification of earlier limed objects may cause lower LAL concentrations due to lasting effects of the liming itself. This effect may last for day/weeks after liming has stopped in running waters, from months to years in lakes and for many years in some terrestrial limed objects.
(2) A generally lower concentration of LAL compared to prelimed concentrations are expected since the amount of acidifying compounds in precipitation has decreased significantly during the last years. This has caused a significant reduction in LAL in most acidified surface waters in Norway (Table I). This decrease should also be expected in Swedish surface waters.

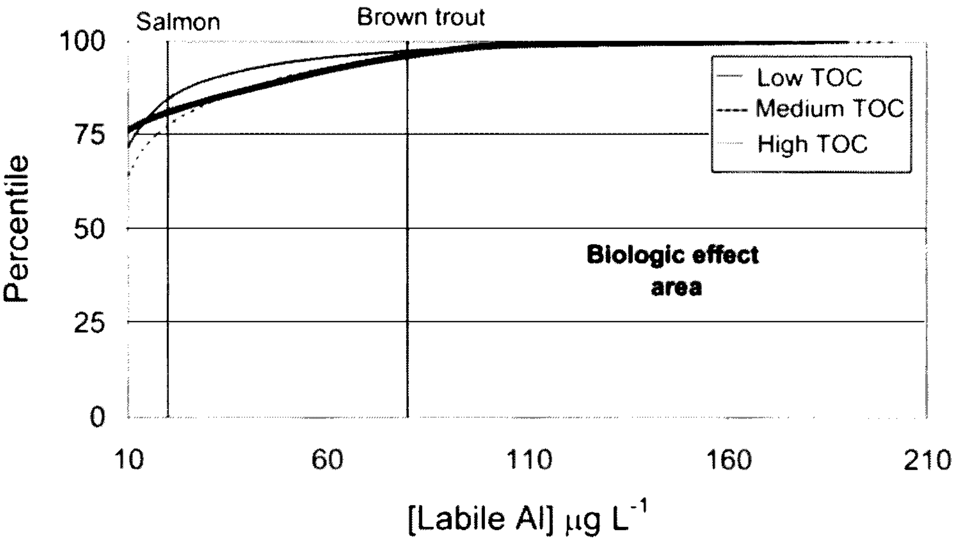

Figure 4. Percentile distribution of labile Al in Norwegian and Swedish lakes based on 3 TOC classes (TOC in mg C L^{-1}). Data are from the Nordic Lake survey 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999). TOC < 5, n = 849; 5 ≤ TOC < 10, n = 459; TOC ≥ 10, n = 219.

However, there is no doubt that reacidification of earlier limed water bodies and catchments might cause negative effects on the aquatic life, because the LAL-concentration in surface waters is rather pH-sensitive within the pH-range: 4.0–5.5. Thus, according to Table VI, reacidified pH should normally not be lower than 5.4, except for high TOC lakes, to avoid large ecological effects on fish.

4.4. CADMIUM

The main source of Cd to surface waters is atmospheric inputs, either indirectly from the catchment or directly to the lake surface. In acid lakes, higher concentrations of Cd appear because of decreased sedimentation. Because the retention time for Cd normally is relatively short, particularly in acidic systems, atmospheric deposition is critical for the lake concentration (Lydersen and Löfgren, 2000). The atmospheric inputs of Cd have decreased in most areas of Scandinavia during the last years. In Norway, Cd in wet-deposition has decreased 50–80% from 1978 to 1996 (Tørseth and Manø, 1997) and in Sweden 25–53% from 1984 to 1996.

By dividing the 1995 surveyed Nordic lakes into TOC- and pH-classes, it is clear that pH plays and important role for the concentration of Cd in surface water (Figure 8), most likely due to the link between high atmospheric inputs of strong acids and Cd and low surface water pH. For Norwegian and Swedish lakes with pH < 5.4, median Cd concentration was 0.030 μg Cd L^{-1}, while the corresponding concentrations in pH-class 5.4–6.0 and pH-class ≥ 6 are 0.014 and 0.010 μg Cd L^{-1}, respectively.

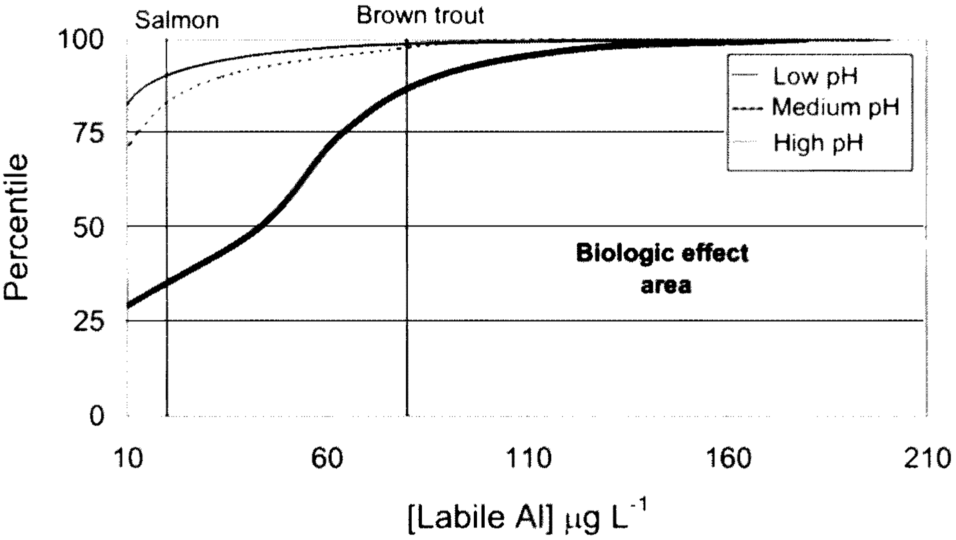

Figure 5. Percentile distribution of labile Al in Norwegian and Swedish lakes based on 3 pH classes. Data are from the Nordic Lake survey 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999). pH < 5.4, $n = 242$; $5.4 \leq$ pH < 6.0, $n = 199$; pH ≥ 6, $n = 1083$.

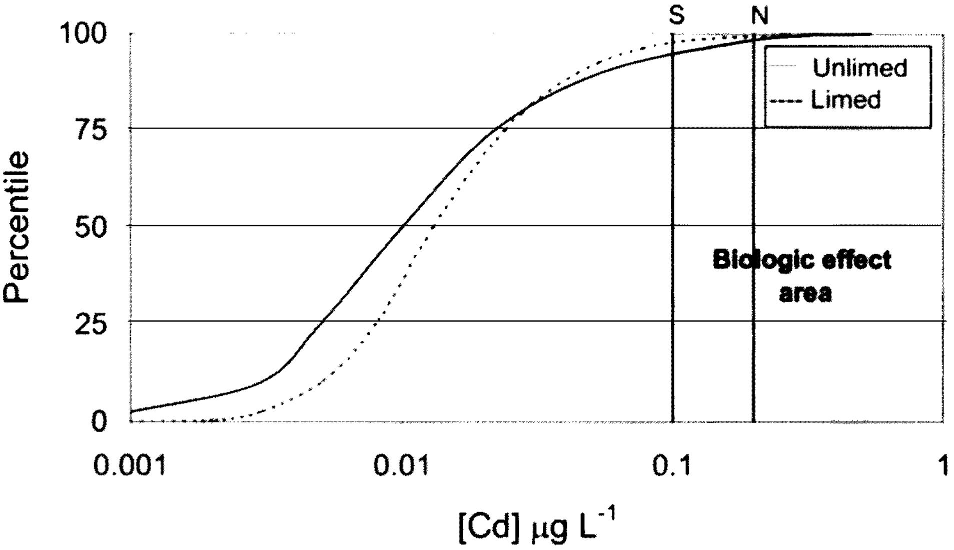

Figure 6. Percentile distribution of total Cd in limed ($n = 261$) and not limed ($n = 820$) Swedish lakes. S and N show the assessed critical concentrations for aquatic life in Norway and Sweden. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

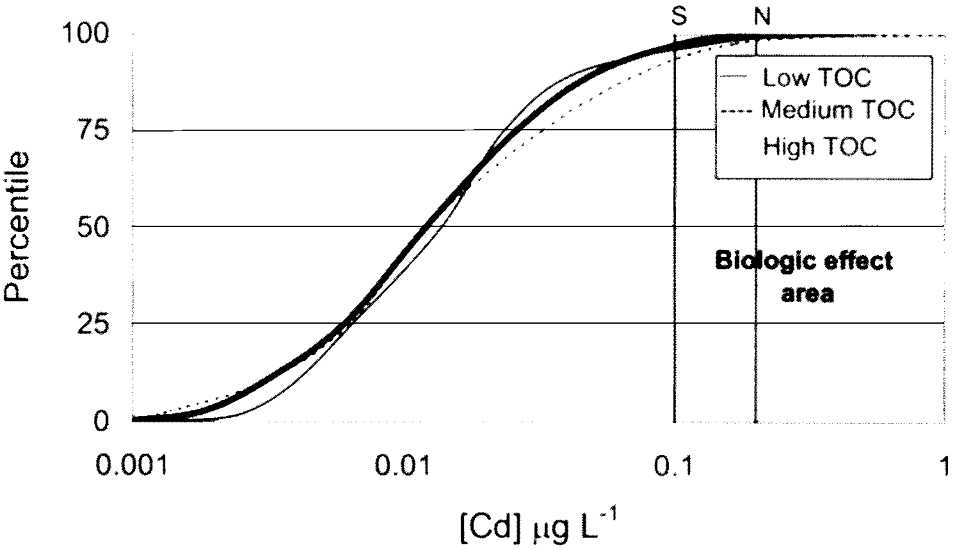

Figure 7. Percentile distribution of total Cd in Norwegian and Swedish lakes based on 3 TOC classes (TOC in mg C L^{-1}). TOC < 5, n = 915; 5 ≤ TOC < 10, n = 427; TOC ≥ 10, n = 202. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

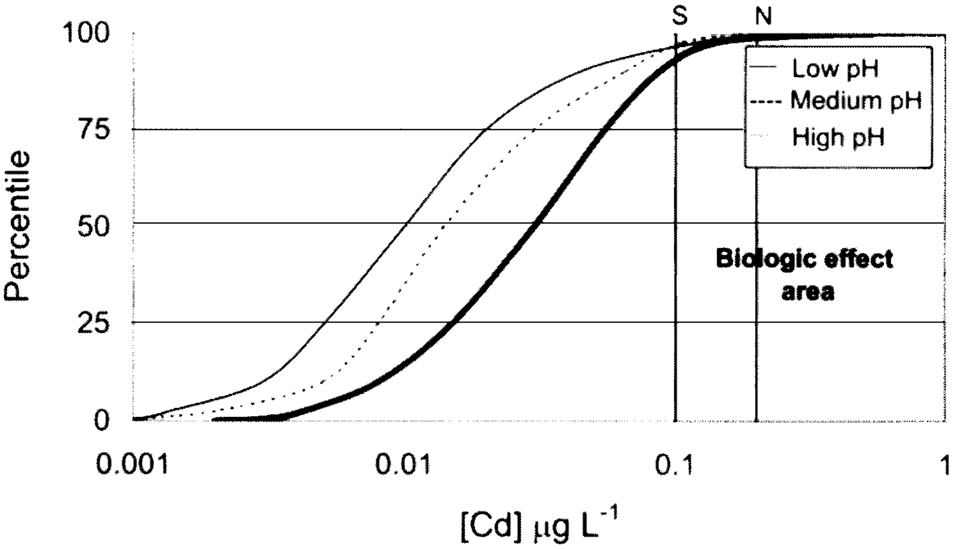

Figure 8. Percentile distribution of total Cd in Norwegian and Swedish lakes based on 3 pH classes. pH < 5.4, n = 250; 5.4 ≤ pH < 6.0, n = 204; pH ≥ 6, n = 1090. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

The concentration of TOC in lakes seems to have little or no effect on the Cd-concentration in surface waters (Figure 7). As for Zn, local geology is essential for high Cd concentration in some surface waters.

The main toxic form of Cd is the aqueous free metal ion, Cd^{2+}. This ion is the only important inorganic species across the pH range of interest: 4.5–7.0. Cadmium toxicity is markedly reduced in presence of organic complexing ligands (Wren and Stevenson, 1991). Cd-concentrations $<0.1\ \mu g\ L^{-1}$ are not considered to be harmful to aquatic organisms, but values between $0.1–1.0\ \mu g\ L^{-1}$ may disturb reproduction for certain species, and Cd $>1\ \mu g\ L^{-1}$ is toxic for several aquatic organisms (Eisler, 1985; Spry and Wiener, 1991; Wren and Stephenson, 1991; Lydersen and Löfgren, 2000).

The lowest biological risk level (LBRL) for Cd in surface water in Norway is $0.2\ \mu g$ Cd L^{-1} (SFT, 1997), while the corresponding LBRL-value in Sweden is $0.1\ \mu g$ Cd L^{-1} (Alm *et al.*, 1998). Based on the regional survey in 1995 (Skjelkvåle *et al.*, 1999), the median concentrations of Cd in Norway, Sweden and Finland are 0.014, 0.010 and $0.011\ \mu g$ Cd L^{-1}, respectively. Because Norway has the highest number of acidic lakes and also the largest area of strong acid rain impact, the highest Cd-concentrations should also be expected in the Norwegian lakes. The median concentration in limed Swedish lakes is $0.013\ \mu g$ Cd L^{-1} (Figure 6). This is somewhat higher than non-limed Swedish lakes. This is likely because many of the limed lakes were relatively acidic before limed. According to the 1995 Nordic Lake Survey, most lakes in Sweden and Norway have Cd-concentrations below the LBRL-values of the two countries (Table V).

There is no good evidence for increased Cd in lakes after a reacidification of limed objects, if reacidified pH of surface water is not lower than pH 5.0–5.5 and the atmospheric inputs of Cd has not increased. Many lakes in Norway and Sweden are relatively close to the LBRL-value of $0.1\ \mu g$ Cd L^{-1}, indicating that minor increases in Cd-concentrations in these lakes may have biological consequences. On the other side, based on the literature review by Lydersen and Löfgren (2000), several studies indicate a certain adaptation to a certain Cd level as well as increased tolerance for Cd at lower pH compared with higher. Thus, if a certain increase in Cd occurs during reacidification of limed waters, the higher Cd tolerance at lower pH may compensate for this increase.

4.5. LEAD

The main source of Pb to surface water systems is atmospheric inputs, both to the catchment and directly on the lake surface. Due to the strong association of Pb with natural organic compounds, Pb is primarily transported to surface waters as Pb-organic complexes. Within normal pH-values of surface waters, pH has minor effects on both transport of Pb to surface waters and sedimentation of Pb in lakes. Accordingly the most important factor for the Pb-concentrations in surface waters are the atmospheric inputs, an input which has decreased significantly during the

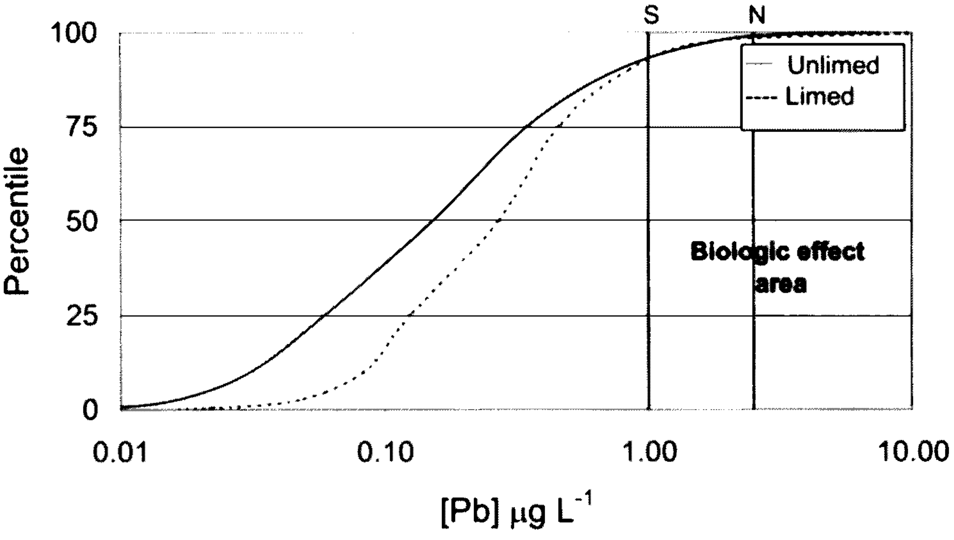

Figure 9. Percentile distribution of total Pb in limed (n = 261) and not limed (n = 820) Swedish lakes. S and N show the assessed critical concentrations for aquatic life in Norway and Sweden. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

last years. In Norway, Pb in wet deposition has decreased by 60–80% from 1978 to 1996 (Tørseth and Manø, 1997) and in Sweden by 48–76% from 1984 to 1996. Already in 1985 Renberg reported decreased Pb concentrations in Swedish lake sediments, as a direct effect of large reduction of Pb as gasoline antiknock additives during the last decade.

Data from the Swedish and Norwegian lake surveys in 1995 (Skjelkvåle *et al.*, 1999) show relatively clear relationships between Pb-concentrations in surface waters, TOC, and pH (Figures 10 and 11). The median concentration of Pb in the pH-class ($pH < 5.4$) was 0.66 μg Pb L^{-1}, while in the pH-class (pH: 5.4–6.0) and pH-class ($pH \geq 6.0$), median Pb concentrations were 0.27 and 0.11 μg Pb L^{-1}, respectively. Dividing the Swedish and Norwegian lakes in TOC-classes, the median Pb-concentration in the TOC-class ($TOC < 5$ mg C L^{-1}) was 0.13 μg Pb L^{-1}, while the TOC-class (TOC: 5–10 mg C L^{-1}) and TOC-class ($TOC \geq 10$ mg C L^{-1}) had median values of 0.17 and 0.32 μg Pb L^{-1}, respectively. The positive correlation between TOC and Pb was expected, but the even more distinct relationship between pH and Pb was not expected, based on pure chemical evaluations only. We assume the strong pH-relationship with Pb is because of the very close link between atmospheric inputs of strong acids and Pb. Thus, it is difficult to distinguish the pH effect from the atmospheric input.

Limed lakes in Sweden normally have higher Pb-concentrations than non-limed Swedish lakes, i.e. median Pb-concentration in limed lakes is 0.27 μg Pb L^{-1} compared with 0.15 μg Pb L^{-1} in non-limed Swedish lakes (Figure 9). This indicates that liming is generally not very effective for precipitation/retention of many heavy

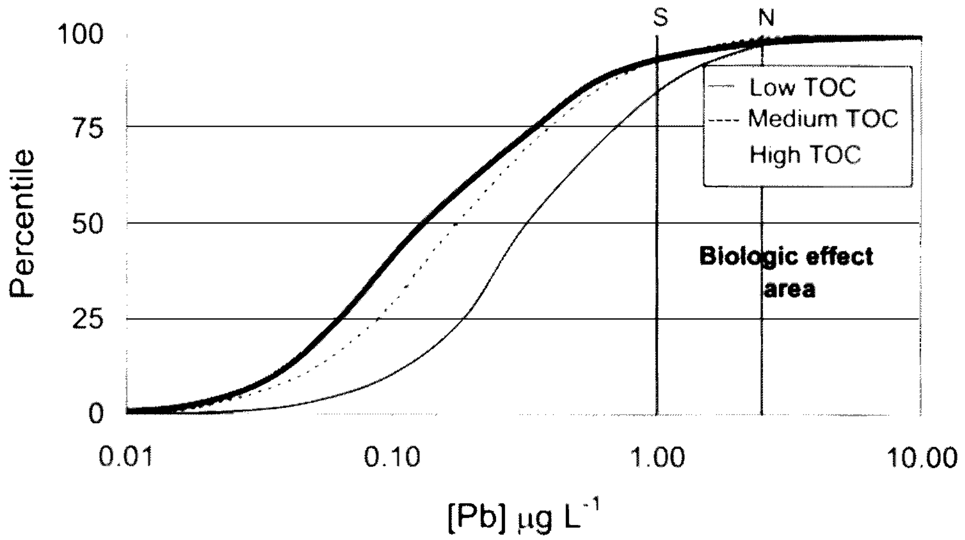

Figure 10. Percentile distribution of total Pb in Norwegian and Swedish lakes based on 3 TOC classes (TOC in mg C L^{-1}). TOC < 5, n = 915; 5 ≤ TOC < 10, n = 427; TOC ≥ 10, n = 202. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

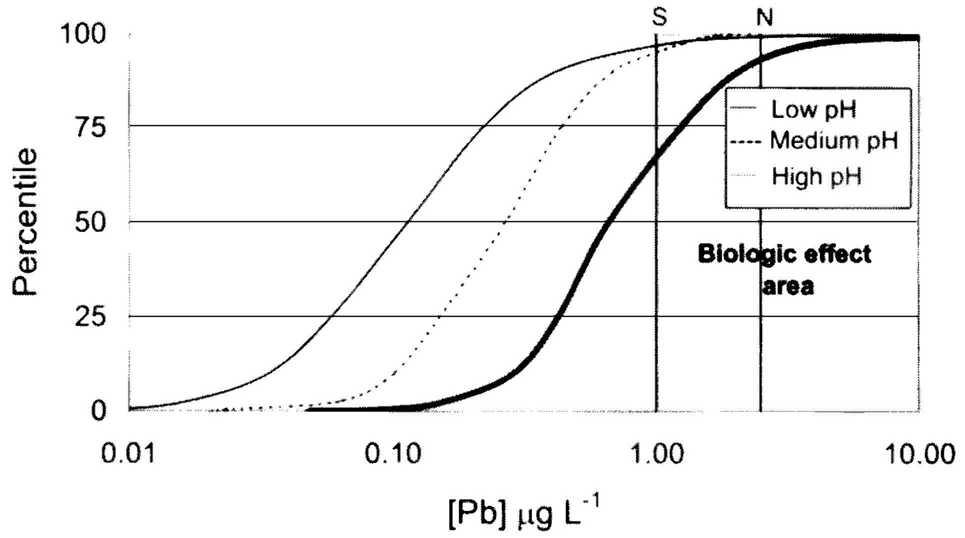

Figure 11. Percentile distribution of total Pb in Norwegian and Swedish lakes based on 3 pH classes. pH < 5.4, n = 250; 5.4 ≤ pH < 6.0, n = 204; pH ≥ 6, n = 1090. Data are from the Nordic Lake Survey, 1995 (Henriksen *et al.*, 1998; Skjelkvåle *et al.*, 1996, 1999).

metals, especially for metals with strong complexing properties to organics such as Pb.

The most bioavailable or most acutely toxic inorganic forms of Pb in slightly acidic freshwaters are Pb^{2+}, $PbCl^{+}$ and $PbOH^{+}$, but some organic Pb-complexes such as tetraethyl- and tetramethyl-Pb are acutely toxic (Eisler, 1988). The toxicity of Pb complexed to natural organic acids has not been examined. We assume that Pb associated with organic acids are less toxic or even non-toxic to aquatic life, a common situation for many other heavy metals (Lydersen and Löfgren, 2000).

Biological effects of Pb are reported to occur at concentration between 5–10 μg Pb L^{-1} (Eisler, 1988; Spry and Wiener, 1991; Wren and Stephenson, 1991; Lydersen and Löfgren, 2000) in low alkalinity waters. The lowest biological risk level (LBRL) for Pb in surface water in Norway is 2.5 μg Pb L^{-1} (SFT, 1997), while the corresponding LBRL-value in Sweden is 1 μg Pb L^{-1} (Alm *et al.*, 1998).

Based on the toxicity criteria in Sweden and Norway, only a few lakes have Pb-concentrations that might have negative effects on aquatic life. The Nordic Lake Survey also documents that even though several of the limed Swedish lakes are under reacidification they do not show any significant increase in Pb. Thus, it is unlikely that significant changes in the concentration of Pb will occur in limed lakes with reacidified pH > 5.

5. Conclusions on Biological Risks of Metals

Based on the literature study and compared to the preliming period, it us unlikely that enhanced remobilization of LAL or other toxic metals from the catchment, the lake sediment and/or the streambed will occur when limed waters reacidify. Rather, the concentrations in surface waters are expected to be lower than before liming started, because of reduced atmospheric inputs of both strong acids and metals as Cd, Pb, Hg and Zn during the last 10–20 yr. The high alkalinity and pH in many Swedish lakes before liming was initiated (Table II) indicates that the number of waters with high concentrations of heavy metals should be relatively limited. This conclusion is supported by the Nordic Lake Survey in 1995.

The concentrations in lakes relative to the biological effect levels, as well as the chemical properties suggest that the potential biological risks associated to reacidification of limed lakes decrease in the order Al $>>$ Cd $>$ Pb in Norwegian and Swedish freshwaters. The risks associated with Cu, Cr, Fe, Mn, Ni, and Zn are very low and do not obviously have to be considered, since such waters are very rare (<2%). Arsenic (As) is present as anions in surface waters, and accordingly differs significantly from the cationic metals. Only a few lakes have high As-concentration in relation to critical levels in Sweden. High As concentrations are mainly related to agricultural areas and consequently high pH waters.

Aluminium has the largest potential to cause biological damage after reacidification of limed waters. There is no difference in risk assessment between running waters and lakes except that the highest Al concentrations are expected in small headwater streams and lakes. We might also expect temporarily higher concentrations of toxic heavy metal species in headwaters, but the same ranking of metals with respect to toxicity is expected regardless of time period and system. The risk with Al is mainly coupled to the pH decrease and less to the liming technique used to maintain circumneutral pH. Terrestrial liming has the potential to reduce the Al load from soils, but as yet the technique has been used in very restricted areas. Aluminium is therefore the metal, which should set the limit for judging the risk of biological damage due to reacidification of limed surface waters. A plan for cessation of liming should use fish sensitivity to Al as a strategy to reduce the biological damage caused by reacidification. Such a strategy would reduce the biological damage on most other organisms as well.

References

Abrahamsson, I.: 1995, 'Kalkning av våtmarker – metoder och effekter', (Liming of wetlands – methods and effects, in Swedish) *Swedish Environm. Prot. Agency report 4484*, 62 pp.

Alm, G., Bengtson, M., Borg, H., Göthberg, A., Johansson, K., Lindeström, L. and Lithner, G.: 1998, 'Metaller. Bakgrundsdokument till bedömningsgrunder för sjöar och vattendrag', ('Metals. Background document for evaluation criteria of lakes and running waters', in Swedish). *Swedish Environm. Prot. Agency, Prel. report*, Version 98-04-21.

Baker, J. P. and Schofield, C. L.: 1982, 'Aluminium toxicity to fish in acidic waters'. *Water, Air, Soil Pollut.* **18**, 289–309.

Belzile, N. and Morris, J. R.: 1995, 'Lake Sediments: Sources or Sinks of Industrially Mobilized Elements', in J. M. Gunn '(ed.), *Restoration and Recovery of an Industrial Region*, Springer-Verlag, New York, pp. 183–194.

Berge, E.: 1997, *MSC-W Status Report 1997*, Part 1: Emissions, dispersion and trends of acidifying and eutrophying agents. Part 2: Numerical Addendum.

Bulger, A., Lien, L., Cosby, B. J. and Henriksen, A.: 1993, 'Brown trout (*Salmo trutta*) status and water chemistry from the Norwegian Thousand Lake Survey: statistical analysis'. *Can. J. Fish Aquat. Sci.* **50**, 575–585.

Cleveland, L., Little, E. E., Hamilton, S. J., Buckler, D. R. and Hunn, J. B.: 1986, 'Interactive toxicity of aluminum and acidity to early life stages of brook trout'. *Trans. Am Fish. Soc.* **115**, 610–620.

Cronan, C. S. and Schofield, C. L.: 1979, 'Aluminum leaching response to acid precipitation: Effects on high-elevation watersheds in the Northeast'. *Science* **204**, 304–306.

Dalziel, T. R. K., Kroglund, F., Lien, L. and Rosseland, B. O.: 1995, 'Then REFISH (Restoring Endangered Fish In Stressed Habitats) PROJECT, 1988–1994'. *Water, Air, Soil Pollut.* **85**, 321–326.

Davis, A. O., Galloway, J. N. and Nordstrom, D. K.: 1982, 'Lake acidification: Its effects on lead sediment of two Adirondack lakes'. *Limnol. Oceanogr.* **27**, 163–167.

Dickson, W.: 1980, 'Properties of Acidified Waters', in D. Drabløs and A. Tollan (eds.), *Ecological Impacts of Acid Precipitation*, SNSF-project, Proc. Int. Conf. Sandefjord, Norway, pp. 75–83.

Dickson, W.: 1983, 'Liming toxicity of aluminium to fish'. *Vatten* **39**, 400–404.

Driscoll, C. T., Cirmo, C. P., Fahey, T. J., Blette, V. L., Bukaveckas, P. A., Burns, D. A., Gubala, C. P., Leopold, D. J., Newton, R. M., Raynal, D. J., Schofield, C. L., Yavitt, J. B. and Porcella, D. B.: 1996, 'The experimental watershed liming study: Comparison of lake and watershed neutralisation strategies'. *Biogeochem.* **32**, 143–174.

Eisler, R.: 1985, 'Cadmium hazards to fish, wildlife, and invertebrates: A synoptic review', *U.S. Department of the Interior, Fish and Wildlife Service, Biological Report* 85 (1.2), 46 pp.

Eisler, R.: 1988, 'Lead hazards to fish, wildlife, and invertebrates: A synoptic review'. *U.S. Department of the Interior, Fish and Wildlife Service, Biological Report* 85 (1.14), 135 pp.

Hem, J. D. and Roberson, C. E.: 1967, 'Form and stability of aluminum hydroxide complexes in dilute solutions'. *Geol. Survey Water-Supply Paper, 1827-A*, U.S. Government Printing Office, Washington D.C., 55 pp.

Hem, J. D. and Roberson, C. E.: 1990. 'Aluminum Hydrolysis Reactions and Products in Mildy Acidic Aqueous Systems', in D. C. Melchior and R. L. Bassettt (eds.), *Chemical Modeling of Aqueous Systems II*, American Chem. Soc., Symp. Series 416, Washington, DC, Mample Press, York, PA, pp. 429–446.

Hem, J. D., Roberson, C. E., Lind, C. J. and Polzer, W. L.: 1973, 'Chemical interactions of aluminium with aqueous silica at 25 °C'. *Geol. Survey Water-Supply Paper, 1827-E*, U.S. Government Printing Office, Washington D.C. 57 pp.

Henriksen, A., Skjelkvåle, B. L., Mannio, J., Wilander, A., Harriman, R., Curtis, C., Jensen, J. P., Fjeld, E. and Moiseenko, T.: 1998, 'Northern European Lake Survey – 1995. Finland, Norway, Sweden, Denmark, Russian Kola, Russian Karelia, Scotland and Wales'. *Ambio* **27**, 80–91.

Hindar, A. and Lydersen, E.: 1995, 'Er utfelt sedimentert aluminium etter vassdragskalking et mulig miljøproblem', ('Is precipitated aluminium after watercourse liming a potential environmental problem', in Norwegian). *NIVA-report*, LNR-3202, 22 pp.

Hindar, A., Kroglund, F., Lydersen, E., Skiple, A. and Høgberget, R.: 1996, 'Liming of wetlands in the acidified Lake RŻynelandsvatn catchment in southern Norway: Effects on stream water chemistry', *Can. J. Fish. Aquatic Sci.* **53**, 985–993.

LaZerte, B.: 1986, 'Metals and acidification: An overview'. *Water, Air, Soil Pollut.* **31**, 569–576.

Leivestad, H., Jensen, E., Kjartansson, H. and Xingfu, L.: 1987, 'Aqueous speciation of aluminium and toxic effects on Atlantic salmon'. *Annls. Soc. R. Zool., Belg.* **117**(1), 387–398.

Lien, L., Raddum, G. G., Fjellheim, A. and Henriksen, A.: 1996, 'A critical limit for acid neutralizing capacity in Norwegian surface waters, based on new analyses of fish and invertebrate responses'. *Sci. Tot. Environ.* **177**, 173–193.

Lind, C. J. and Hem, J. D.: 1975, 'Effects of organic solutes on chemical reactions of aluminum'. *Geol. Survey Water-Supply Paper, 1827-G*, U.S. Government Printing Office, Washington D.C., 83 pp.

Lydersen, E.: 1992, 'it Aluminium in dilute acidic freshwaters – Chemical, analytical and biological relevance', *Ph.D. Thesis*, Department of Limnology, University of Oslo, 133 pp. + Appendix.

Lydersen, E., Poléo, A. B. S., Nandrup Pettersen, M., Riise, G., Salbu, B., Kroglund, F. and Rosseland, B. O.: 1994a, 'The importance of 'in situ' measurements to relate toxicity and chemistry in dynamic aluminium freshwater systems'. *J. Ecol. Chem.* **3**, 357–265.

Lydersen, E., Fjeld, E. and Andersen, T.: 1994b, 'Fiskestatus og vannkjemi i norske innsjøer' ('Fish-status and water-chemistry in Norwegian lakes', in Norwegian with English summary) *Norwegian Institute for Water Research (NIVA) Report*, O-93172, LNR: 3002–94.

Lydersen, E. and Löfgren: 2000, 'Chemical and biological effects of reacidification of limed water bodies – a state of the art review'. *Swedish Environm. Prot. Agency Report* 5074, 76 pp. [In Swedish]

Mylona, S.: 1996, 'Sulphur dioxide emissions in Europe 1880–1991 and their effect on sulphur concentrations and depositions'. *Tellus* **48 B**, 662–689.

Naturvårdsverket: 1999, *Nationell plan för kalkning av sjöar och vattendrag 2000–2009*. (National plan for liming of lakes and streams 2000–2009, in swedish). Swedish Environm. Prot. Agency, ISSN-7298.

Nelson, W. O. and Campbell, P. G. C.: 1991, 'The effects of acidification on the geochemistry of Al, Cd, Pb and Hg in freshwater environments. A literature review'. *Environm. Pollut.* **71**, 91–130.

Norton, S. A., Hess, C. T. and Davis, R. B.: 1981, 'Rates of Accumulation of Heavy Metals in Pre- and Post-European Sediments in New England Lakes', in S. J. Eisenreich (ed.) *Atmospheric Pollutants in Natural Waters*, Ann Arbor Sci. Publishers, Ann Arbor, pp. 409–421.

Polèo, A. B. S., Østbye, K., Øxnevad, S. A., Andersen, R. A., Heibo, E. and Vøllestad, L. A.: 1997, 'Toxicity of acid aluminium-rich water to seven freshwater fish species: A comparative laboratory study'. *Environm. Pollut.* **96**, 129–139.

Renberg, I.: 1985, 'Influence of acidification on the sediment chemistry of Lake Gärdsjön, SW Sweden'. *Ecol. Bull.* (Stockholm) **37**, 246–250.

Reuss, J. O. and Johnson, D. W.: 1986, *Acid Deposition and the Acidification of Soils and Waters. Ecological Studies*, Vol. 59, Springer, New York, Berlin, Heidelberg, Tokyo.

Schecher, W. D. and Driscoll, C. T.: 1987, 'An evaluation of uncertainty associated with aluminium equilibrium calculations'. *Water Resour. Res.* **23**, 525–534.

Schecher, W. D. and Driscoll, C. T.: 1988, 'An evaluation of the equilibrium calculations within acidification models: The effects of uncertainty in measured chemical compounds'. *Water Resour. Res.* **24**, 533–540.

Schiff, S. L. and Andersson, R. F.: 1986, 'Alkalinity production in epilimnetic sediments: acidic and non-acidic lakes'. *Water, Air, Soil Pollut.* **31**, 941–948.

Schofield, C. L.: 1977, 'Acid snow-melt effects on water quality and fish survival in the Adirondack Mountains of New York State, U.S.', *Research Technical Completion Report* A-072-NY. Office of Water Research and Technology, Department of the Interior, Washington, DC.

Seip, H. M., Andersen, D. O., Christophersen, N., Sullivan, T. J. and Vogt, R. D.: 1989, 'Variations in concentrations of aqueous aluminium and other chemical species during hydrological episodes at Birkenes, southernmost Norway'. *J. Hydrol.* **108**, 387–405.

SFT: 1997, 'Klassifisering av miljøkvalitet i ferskvann' ('Classification of Environmental Quality in Freshwaters', in Norwegian). *The Norwegian Pollution Control Authority (SFT), Veiledning* 97:04, 31 pp.

Skjelkvåle, B. L., Henriksen, A., Faafeng, B., Fjeld, E., Traaen, T. S., Lien, L., Lydersen, E. and Buan, A. K.: 1996, 'Regional Lake Survey 1995. A Water Chemical Survey of 1500 Norwegian Lakes. The National Monitoring Programme for Long-range Transported Air Pollutants'. *Norwegian State Pollution Control Authority, Report* 677/96, 73 pp.

Skjelkvåle, B. L., Mannio, J., Wilander, A., Johansson, K., Jensen, J. P., Moiseenko, T., Fjeld, E., Andersen, T., Vuorenmaa, J. and Røyseth, O.: 1999, 'Heavy Metals in Nordic Lakes; Harmonized Data for Regional Assessment of Critical Limits'. *Norwegian Institute for Water Research, Report*, No. O-98024, Serial No. 4039–99, 71 pp.

Smith, R. E. and Hem, J. D.: 1972, 'Effect of ageing on aluminum hydroxide complexes in dilute aqueous solutions'. *Geol. Survey Water-Supply Paper*, 1827-D, U.S. Government Printing Office, Washington D.C. 51 pp.

Spry, D. J. and Wiener J. G.: 1991, 'Metal bioavailability and toxicity to fish in low-alkalinity lakes: A critical review'. *Environm. Pollut.* **71**, 243–304.

Staurnes, M., Blix, P. and Reite, O. B.: 1993, 'Effects of acid water and aluminum on parr-smolt transformation and seawater tolerance in Atlantic salmon, *Salmo salar*'. *Can. J. Fish. Aquat. Sci.* **50**, 1816–1827.

Staurnes, M., Kroglund, F. and Rosseland, B. O.: 1995, 'Water quality requirement of Atlantic salmon (*Salmo salar*) in water undergoing acidification or liming in Norway'. *Water, Air, Soil Pollut.* **85**, 347–352.

Svensson, T., Dickson, W., Hellberg, J., Moberg, G. and Munthe, N.: 1995, 'The Swedish liming programme'. *Water, Air, Soil Pollut.* **85**, 1003–1008.

ter Braak, C. J. F.: 1994, 'Canonical community ordination. I. Basic theory and linear models'. *Ecoscience* **1**, 127–140.

Thörnelöf, E. and Degerman, E.: 1991, 'Lake Liming in Sweden – Case Studies and General Experience', in H. Olem, R. K. Schreiber, R. W. Brocksen and D. B. Porcella (eds.), *International Lake and Watershed Liming Practices*, The Terrene Inst., Inc., Washington, D.C., pp. 41–60.

Tørseth, K. and Manø, S.: 1997, *Monitoring of Long-range Transported Polluted Air and Precipitation* (in Norwegian). Norwegian State Pollution Control Authority (SFT), Oslo, Report 703/97.

Wilander, A.: 1997, 'Referenssjöarnas vattenkemi under 12 år; tillstånd och trender', ('Reference lake water chemistry during 12 years, conditions and trends', in Swedish). *Swedish Environm. Prot. Agency Report* 4652, 79 pp.

Wilander, A., Andersson, P., Borg, H. and Broberg, O.: 1995, 'The Effect of Liming on Water Chemistry', in L. Henriksson and Y. W. Brodin (eds.), *Liming of Acidified Surface Waters. A Swedish Synthesis*, Springer Verlag, Berlin, pp. 125–178.

Wilander, A., Johnson, R., Goedkoop, W. and Lundin, L.: 1998, 'Riksinventeringen 1995. En synoptisk studie av vattenkemi och bottenfauna i svenska sjöar och vattendrag'. ('A Symnoptic Study of Water Chemistry and Bottom Fauna in Swedish Lakes and Running Waters', in Swedish). *Swedish Environm. Prot. Agency Report*, 4813, 191 pp.

Wood, C. M., McDonald, D. G., Booth, C. E., Simons, B. P., Ingersoll, C. G. and Bergman, H. L.: 1988, 'Physiological evidence of acclimation to acid/aluminum stress in adult brook trout (*Salvelinus fontinalis*). 1. Blood composition and net sodium fluxes'. *Can. J. Fish. Aquat. Sci.* **45**, 1587–1596.

Wood, C. M., McDonald, D. G., Ingersoll, C. G., Mount, D. R., Johansson, O. E., Landsberger, S. and Bergman, H. L.: 1990, 'Effects of water acidity, calcium, and aluminum on whole body ions of brook trout (*Salvelinus fontinalis*) continuously exposed from fertilization to swim-up: A study by instrumental neutron activation analysis'. *Can. J. Fish. Aquat. Sci.* **47**, 1593–1603.

Wren, C. D. and Stevenson, G. L.: 1991, 'The effect of acidification on the accumulation and toxicity of metals to freshwater invertebrates'. *Environ. Pollut.* **71**, 205–241.