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Treatment of lakes and storage reservoirs with very low dosages of selenium to reduce methyl mercury in fish

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Treatment of lakes and storage reservoirs with very low dosages of selenium to reduce methyl mercury in fish

Sammanfattning/Summary

The trials of the addition of selenium to three lakes indicate a rapid bioackumulation of selenium. The selenium concentrations in zooplankton increased, and these constitute the first and most important level in the aquatic ecosystem. The increase of selenium in zooplankton resulted in a reduction in the concentration of methyl mercury. The bio-magnification in the food chain led to increases in selenium concentrations and reductions in methyl mercury. This in turn led to reduced concentrations of mercury in crayfish and methyl mercury in younger fish. After a certain delay, the bio-magnification of selenium continued in the higher trophic levels. This resulted in increased selenium concentrations and reduced methyl mercury concentrations in older perch and pike several years after the zooplankton, young roach and young perch. When the selenium treatments stopped the concentrations of selenium in the water, aquatic moss, bentic invertebrates and the youngest age groups of roach and perch fell immediately. The reductions in selenium in these parts of the ecosystem are related to the water retention period of the lake or reservoir.

Nyckelord samt ev. anknytning till geografiskt område eller näringsgren /Keywords

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Innehållsförteckning

1 Introduction

1.1 The effects of damming on methyl mercury and selenium concentrations in fish

High concentrations of methyl mercury in fish have been a significant environmental problem for a long period of time and affect around 50% of Sweden's lakes, primarily nutrient-poor woodland lakes. Another problem is the increased concentration of methyl mercury in fish in storage reservoirs, and in lakes downstream of these reservoirs, following the flooding of what were previously woodland and wetland areas. This is caused by mercury and methyl mercury, which had accumulated in organic material in the soil, leaching out of the soil profile. The larger the area dammed per volume unit, the greater the leaching effect.

These are well-known effects and have previously been observed in storage reservoirs in countries such as Finland, the USA and northern Canada. In Sweden there are many examples of storage reservoirs in forests and wetlands. The Skinnmuddselet is a reservoir in Västerbotten/Västernorrland, which after having been in existence for more than 13 years now has significantly increased methyl mercury concentrations in pike, older perch and burbot. The high concentrations in the fish are caused by methyl mercury seeping out of forest soil, peat and floating peat.

It has recently emerged that there are lower concentrations of selenium in the fish and in the water in storage reservoirs in Canada and Sweden than in lakes in comparable areas. One hypothesis is that this effect of damming is caused by the fact that the form and concentration of selenium in water is not the same, at least initially, in a storage reservoir as in a natural lake. Damming to form reservoirs in former forests and wetlands can, for example, result in changes to the redox ratios and/or methylation, which in its turn leads to increases in soil retention of selenium of its discharge as a gas into the atmosphere. This will mean a reduction in the selenium concentrations in the water and the fish. Another cause could be that damming could prevent selenium being converted into forms, which can be absorbed by fish. A further cause could be that damming results in a change from a flowing water system to a lake ecosystem, which affects bioackumulation of selenium in the food chain.

1.2 Background

1.2.1 Mercury and selenium in fish after the damming of the Skinnmuddselet

Experiences from storage reservoirs in countries such as Finland and Canada show that, regardless of the selenium concentrations in fish, the methyl mercury concentrations in young fish decrease significantly during the 10 years after the construction of the dam. The concentrations decrease most quickly in the youngest age groups of roach and perch, and reach levels corresponding to or directly above those measured before the creation of the reservoir within a 5-year period. The methyl mercury concentrations decrease most slowly in older fish-eating species such as pike, burbot and older perch. It often takes between 15 and 20 years, sometimes longer, before the mercury concentrations in these fish fall to the levels seen before the creation of the dam.

The developments in the Skinnmuddselet on the river Gideälven followed the patterns described above after it was dammed in 1989. Methyl mercury in small perch (10 cm long, 0.01 kg and around 2 years old) reached its highest concentrations in 1991, and then decreased by around 50% up to and including 1993. The concentrations in pike weighing 1 kg (around 3-4 years old) increased more slowly than in young perch and reached their highest levels one year later during 1992.

The mercury concentrations in 1-kg pike then decreased during the period from 1993 to 2002, that is from 1.4 mg/kg to 0.9 mg/kg or around the previous level for blacklisting. The mercury concentrations in older perch (25 cm long, 0.2 kg and around 5-7 years old) have decreased in the Skinnmuddselet from the peak of 1.7 mg/kg in 1994 to 1 mg/kg in 2001.

The Stora Tällvattnet, the lake directly downstream from the storage reservoir, has also seen a reduction in the mercury concentrations in larger pike and older perch. The mercury concentration has decreased from 2.4 mg/kg in older perch in 1996 to 1.5 mg/kg in 2001. The methyl mercury concentration in young perch continued to decrease after the peak 1991. The concentration of methyl mercury in young perch 13 years after the construction of the dam was around 80% lower than in 1991, both in the Skinnmuddselet and the Stora Tällvattnet.

The selenium concentration in fish is lower (around 0.1 mg/kg) in the Skinnmuddselet and the Stora Tällvattnet than in nearby lakes, where the concentration is 2 to 3 times higher. The selenium concentration in the water is also lower in the Skinnmuddselet and the Stora Tällvattnet than in other lakes in the area.

1.2.2 Regional studies of selenium and mercury

One predominant source of selenium in the atmosphere is the burning of coal. In Sweden these emissions come primarily from other countries. Crystalline bedrock generally has low natural levels of selenium. Acidified soil profiles also result in even lower release of selenium from the soil to streams and lakes.

Concentrations of selenium in water and fish are generally lower in northern Sweden than they are further south. This is primarily caused by a lower atmospheric deposition of selenium, with dry and wet deposition in northern Sweden and naturally low levels in the bedrock.

Even the naturally low concentrations of selenium found in Lake Water influence mercury concentrations in fish. A regional comparison carried out in Västernorrland shows that there is a clear connection between the higher concentrations of selenium and the lower concentrations of methyl mercury in pike. The natural selenium concentrations in Lake Water in northern Sweden are generally lower than 0.1 µg/l.

Similar results were obtained in a Norwegian study presented at the international mercury conference in Hamburg in 1996. In the study, selenium and mercury concentrations in perch from lakes in southern Norway were analysed.

Even concentrations of selenium as low as 0.3 to 0.4 mg/kg in pike often result in the methyl mercury concentrations being limited to around 0.5 mg/kg. Therefore the very low selenium concentrations found naturally in Lake Water in Västernorrland and Västerbotten should result in low selenium and high mercury concentrations in fish. Damming produces very low selenium concentrations in storage reservoirs, which leads to even higher concentrations of methyl mercury in fish.

1.2.3 Previous full-scale trial of treatment with selenium in Sweden

The following is a summary of the results of a review of unpublished data from low-dosage treatments with selenium as part of an earlier project ("Liming-Mercury-Caesium") carried out by the Swedish Environmental Protection Agency.

It had previously been observed that in lakes in the USA and Canada an increased uptake of selenium in fish resulted in lower concentrations of methyl mercury. The full-scale trial carried out by the Swedish Environmental Protection Agency also showed that the addition of selenium to Lake Water resulted in a significant reduction in concentrations of methyl mercury in fish. In some of the Swedish experiments selenium treatment has, however, resulted in damage to fish stocks. Amongst other things, there have been negative effects on the reproduction of perch. The high dosages resulted in the selenium concentrations in fish in many cases being at the same level as or above the concentrations regarded as acceptable in food.

The experiences from many of the Swedish Environmental Protection Agency's earlier trials showed that significantly lower dosages of selenium still had a positive effect on the methyl mercury concentrations in fish, without causing harm to invertebrates, plants or fish.

The Swedish Environmental Protection Agency and the county council carried out trials of selenium in Lake Oppsjön and Lake Ödingen during 1987, 1988 and 1989. The highest selenium concentrations in the water were 1 to 1.7 µg/l after the first dose, around 5 µg/l after the second and then 1 to 1.5 µg/l after the third. On the third occasion a slower-release rubber/selenium tube was used. Both Lake Oppsjön and Lake Ödingen contain around 9 million cubic metres of water and have a water retention period of 0.68 and 1.1 years respectively. In both cases the selenium concentrations in young perch increased immediately and corresponded to the increase in selenium in the water. A year after the last treatment, the concentrations had fallen quickly again in the young pike corresponding to the rapid reduction in selenium concentrations in the water. The mercury concentrations in perch decreased in both Lake Oppsjön and Lake Ödingen in relation to the increase in the selenium concentrations in the fish. The selenium concentrations in young perch during 1995, six years after the last treatment, were 1.3 and 1.1 mg/kg respectively, that is around four times higher than before the treatment. The mercury concentrations in young perch in 1995 were lower than in 1987, before the treatments.

The process for pike was somewhat different to that for young perch. The concentration of selenium increased more slowly and the mercury concentration decreased more slowly. The selenium concentrations reached a maximum in pike during 1992 and decreased only slowly during the following years. The same developments were observed in both lakes. The most noteworthy feature is that the mercury concentration in pike did not increase between 1993 and 1996 while the selenium concentration was falling. Instead it continued to decrease during the whole period of the trial. During 1995 and 1996 the methyl mercury concentration in pike was 0.3 to 0.5 mg/kg or 4 to 6 times lower than in the years before the addition of selenium.

The similar reduction in methyl mercury concentrations in pike in Lake Oppsjön and Lake Ödingen is even more noteworthy because Lake Ödingen was dammed in 1989 to turn it into a storage reservoir for hydroelectric power. This involved flooding forests and wetlands, which in the past has always resulted in large increases in the concentration of methyl mercury in fish. One explanation could be that the earlier treatment with selenium counteracted this effect and the increase in the mercury concentration in fish as a result of flooding did not occur. Another could be that the area flooded was significantly smaller than in the Skinnmuddselet and that therefore the increase in mercury did not take place.

During the course of this project, water samples were taken again from Lake Oppsjön and Lake Ödingen and analysed for Hg-tot, MeHg and Se. These analyses show that the concentrations of Hg-tot and MeHg are currently similar to the concentrations in the Skinnmuddselet storage reservoir and the lakes downstream of it, while the concentration of selenium in Lake Ödingen is around 4 times higher (around 0.2 µg/l). This selenium concentration is thought to be sufficient to protect the fish stocks in the long term from a significant increase in methyl mercury. The same is true of Lake Oppsjön, which has a concentration of selenium of around 0.1µg/l in the water.

The continued tracking of the trial in Lake Oppsjön and Lake Ödingen, the storage reservoir, shows that a low dosage of selenium can result in a significant reduction in methyl mercury concentrations in perch and pike **over several years**. The addition of selenium did not have any demonstrable negative effects on other parts of the ecosystem. Fish from the treated water was regularly used for human consumption.

Analyses of aquatic moss showed a twofold to fourfold increase in the selenium concentration in Lake Oppsjön and Lake Ödingen 10 years after the treatments with selenium. The lake sediment and the animals living on the bottom of the lake, such as *Asselus*, showed a twofold increase in selenium concentrations, but the analyses of pondweed showed low selenium concentrations after the treatments.

Treatments with mixed lime (selenium and lime) produced estimated maximum increases in the selenium concentration in the water of around 0.02 – 0.43 µg/l. The mercury concentration reduced by between 8 and 53% in perch and 0 and 43% in pike in the different lakes. This shows that significant reductions in methyl mercury concentrations in fish can be achieved with very low doses of selenium.

Three of the most important results of the literature review carried out in the project (Parkman and Hultberg) show that:

1. selenium concentrations > 1µg/l in water can cause damage at the lower trophic levels in a lake's ecosystem (primarily to zooplankton)
2. both selenium and methyl mercury increase in concentration through bio-accumulation via the food chain
3. No effects have been observed in fish with concentrations, which are < 3 µg/l.

An examination study was produced (Fredriksson, R.) in cooperation between the project and the University of Kalmar. In the course of toxicology tests carried out on invertebrates both in the field and in the laboratory, no mortality was observed. The selenium concentrations used in this trial ranged from those corresponding to concentrations in the selenium treatments in Lake Lavsjön and other lakes right up to concentrations several orders of magnitude higher.

The information given above shows that a relatively long-term reduction in methyl mercury concentrations in fish can be achieved by very low dosages of selenium in lakes and storage reservoirs. Large parts of the areas currently being treated in Sweden also have a selenium deficiency for natural reasons. Selenium treatment may therefore also have a positive effect on public health by producing fish with lower methyl mercury concentrations and higher selenium concentrations.

HYPOTHESIS

Reservoirs and lakes with a high concentration of methyl mercury in fish can be treated by the addition of very low dosages of selenium, which produces a twofold positive effect, by reducing the methyl mercury concentration in fish and at the same time increasing the selenium concentration.

2 The addition of selenium to lakes Lavsjön, Trehörningen and Vrångevatten

The research project started with the addition of selenium to Lake Lavsjön in Västerbotten during 1998. The work at Lake Lavsjön was carried out in cooperation between IVL, Graninge Kraft AB, the University of Kalmar and Signe Persson from Gothne in Västernorrland.

The selenium treatment was started in two other lakes in southern Sweden in 1999: Trehörningen in Hällefors in the county of Örebro and Vrångevatten in the Herrestadsfjället mountains in Uddevalla in Västra Götaland. Staff at IVL,

Signe Persson from Gothne in Västernorrland and the University of Kalmar worked together on the field trials carried out on these two lakes. The Hällefors fishery conservation department and the Aquaculture College in Sävenfors also took part in the Trehörningen fieldwork. At Lake Vrångevatten the Uddevalla sport-fishing club, which manages the fish stocks in the Uddevalla area, also took part.

Selenium was added to Lake Lavsjön during 1998, 1999 and 2000 and to Lakes Trehörningen and Vrångevatten during 1999 and 2000 with the objective of reaching a level of 0.5 – 1 $\mu g/l^{-1}$.

The selenium treatment in Lake Lavsjön was very labour-intensive, partly because of the lake's naturally short water retention period and partly because of the high levels of precipitation during the period of the trial. The treatment was carried out around every two weeks during the summer in each of the three years.

As a result of the longer retention time in Lakes Trehörningen and Vrångevatten, the treatment was only carried out there once or twice per year.

2.1 Samples

The results of the selenium additions were monitored by means of samples taken from May to October, when the lakes were not covered with ice. The selenium and methyl mercury concentrations in the water, the sediment, the plankton, bentic animals, plants, roach, perch and pike, and other species of fish were analysed. Fish reproduction was monitored by sampling and analysing fry and young fish caught with bag nets and fine-mesh fry nets. The composition of the fish population was studied by catching with a standardised sampling net. In Lake Vrångevatten samples of Swedish crayfish were taken and analysed for selenium and mercury.

3 Results

3.1 Lake data

The areas, water retention period, some variables concerning the water chemistry, total mercury, methyl mercury and selenium in the water, plus methyl mercury in perch and 1 kg pike before the start of the selenium treatment are shown in Table 1.

Table 1	LAVSJÖN	TREHÖRNINGEN	VRÅNGEVATTEN
AREA (ha)	70	6	50
RETENTION TIME (YEARS)	0.1	1.9	1
pH	6.2	6.4	6.6
COLOR (mg/Pt/L)	100	70	30
DOC (mg/l)	12	10	6
CALCIUM (mg/L)	2.2	3.7	5.5
MAGNESIUM (mg/L)	0.50	0.65	0.80
Total Hg in water (ng/L)	3.0	2.8	2.4
MeHg in water (ng/L)	0.23	0.20	0.10
MEAN Se (natural) in water (ng/L)	47	35	70
MEAN MeHg in 1 kg pike (mg/kg ww)	0.8	0.7	0.6
MEAN MeHg (mg/kg ww) in 10 cm perch	0.11	0.08	0.05
MEAN Se in 1 kg pike (mg/kg ww)	0.3	0.1	0.4
MEAN Se (mg/kg ww) in 10 cm perch	0.5	0.4	0.6

3.2 Addition of selenium

Figure 1 shows the amounts of selenium added in the form of sodium selenite.

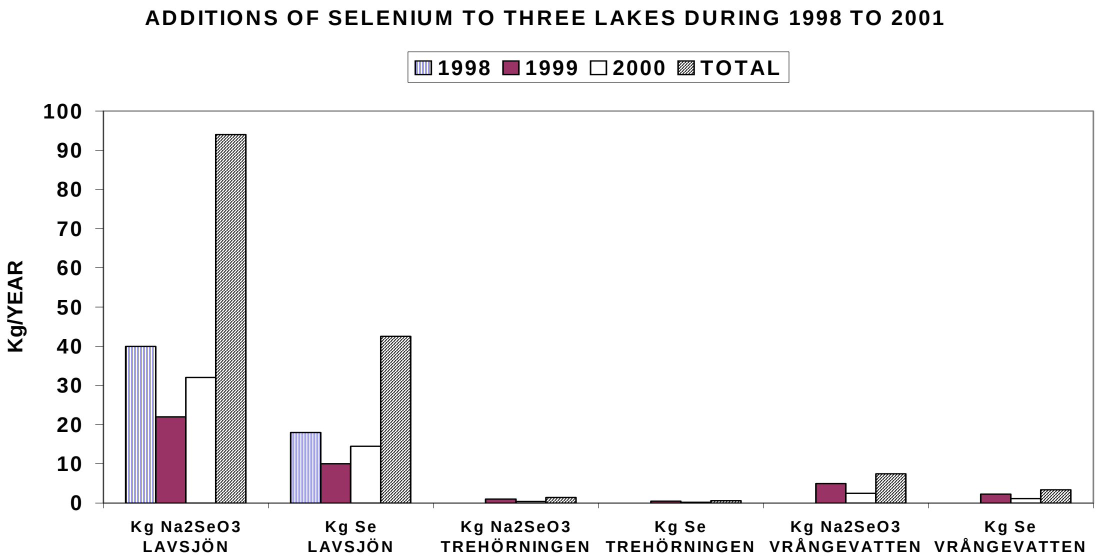

Figure 1

During 1998, 1999 and 2000 a total of 42.5 kg of selenium was added to Lake Lavsjön. During 1999 and 2000 0.6 kg was added to Lake Trehörningen and 3.4 kg to Lake Vrångevatten.

3.3 Selenium concentrations in the treated lakes

The selenium concentration in Lake Lavsjön varied between 0.1 and 1 µg/l. The lowest values occurred directly before the treatment and during the winter. The highest values were obtained at the time of the selenium treatments. In Lakes Trehörningen and Vrångevatten the selenium concentration was kept stable at around 0.5 – 1 µg/l-1 with relatively little work. The selenium concentration in the water in both Lake Trehörningen and Lake Vrångevatten was clearly higher even after the winter of 1999/2000, in contrast to Lake Lavsjön. Because of its short water retention period, Lake Lavsjön had a background concentration of selenium in the water both after the winter of 1998/1999 and the winter of 1999/2000.

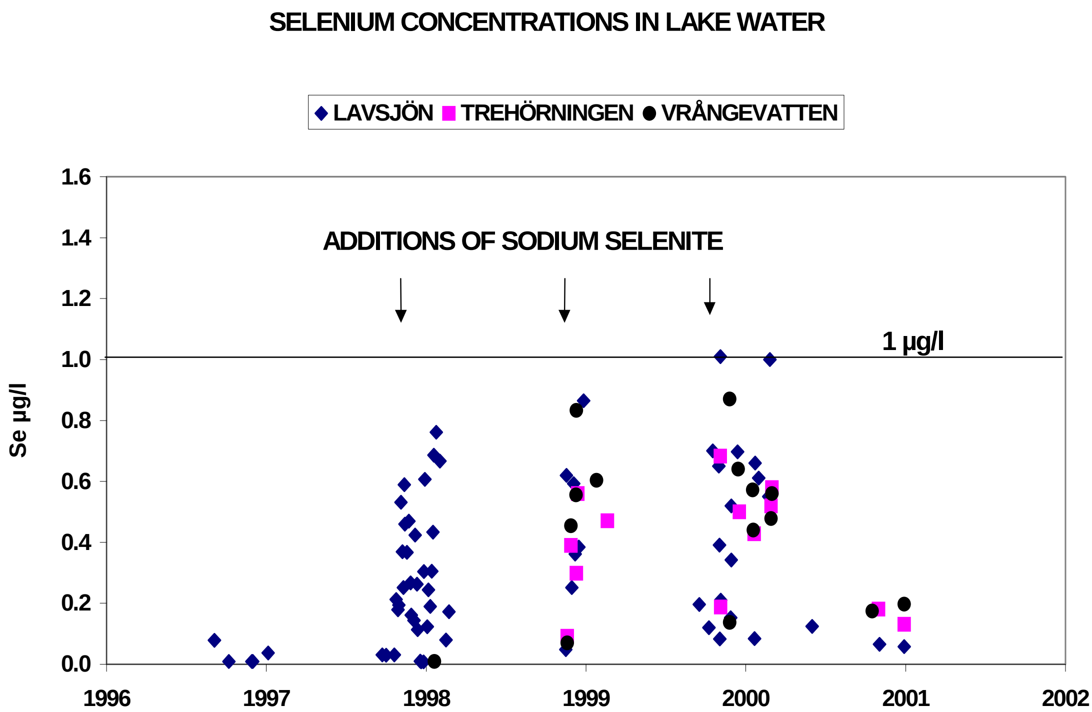

Figure 2

The average concentration in Lake Lavsjön (see Figure 3 below) during the summer months in the year before the treatments was around 0.05 µg Se/l. In the years when selenium was added, this increased to 0.29, 0.42 and 0.46 µg/l and it then fell back to 0.06 µg/l in 2001, when no more selenium was added. In Lake Trehörningen, which had the longest water retention period (1.9 years), the selenium concentration before the treatments was 0.04 µg/l. This increased to 0.36 and 0.48 respectively when the selenium was added and then decreased to 0.16 µg/l in 2001. Lake Vrångevatten had the highest selenium concentrations both before and during the treatments. Before the treatments the concentration was 0.07 µg/l, at the time when the selenium was added it was 0.50 and 0.53 respectively and during 2001 it was 0.19 µg/l.

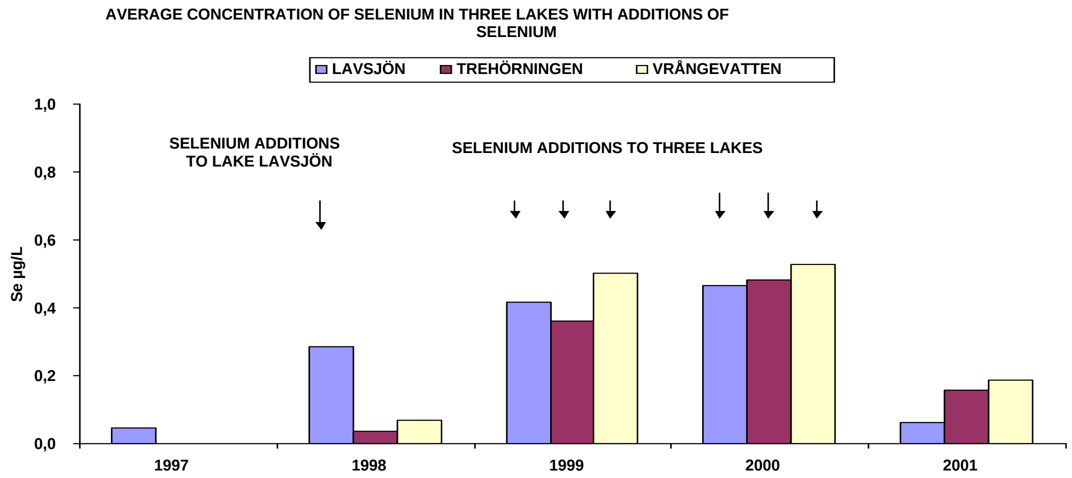

Figure 3

The concentrations of methyl and total mercury in the lake water were not affected by the additions of selenium. This can be seen in Figures 4 and 5 below which use the example of Lake Lavsjön.

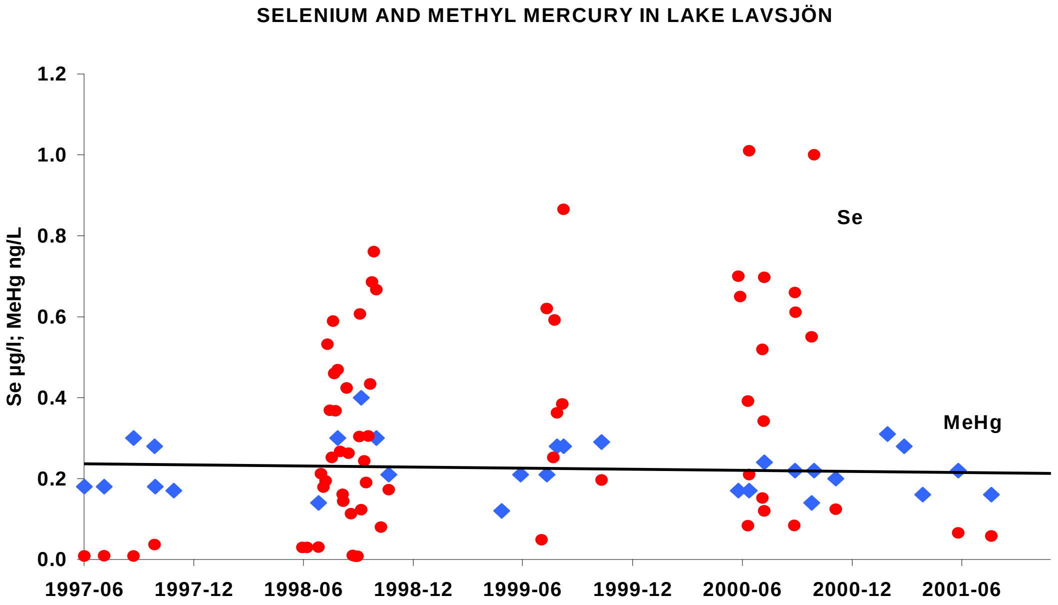

Figure 4

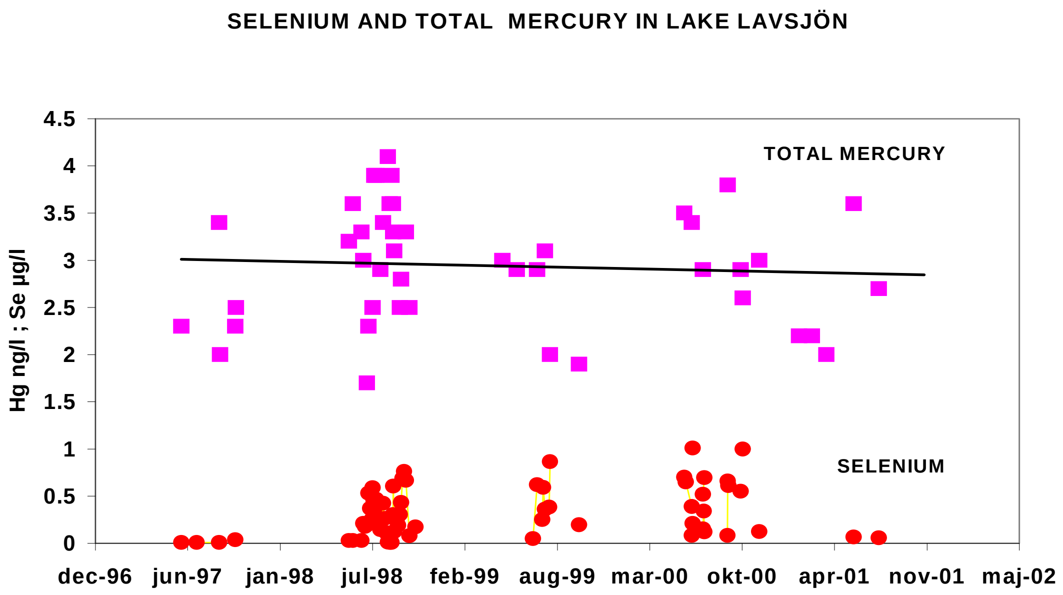

Figure 5

The results of sampling sediment from the bottom of the lake in shallow and deep areas are shown in Figures 6 and 7. It is clear that the selenium concentration increased by a factor of around 2. However, the mercury concentrations have not changed.

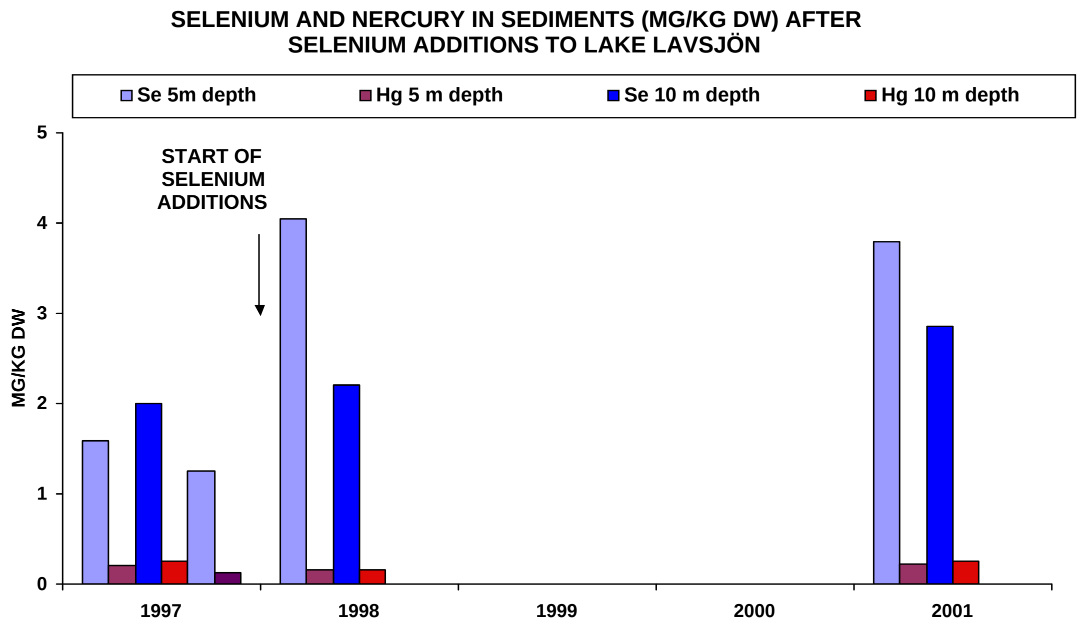

Figure 6

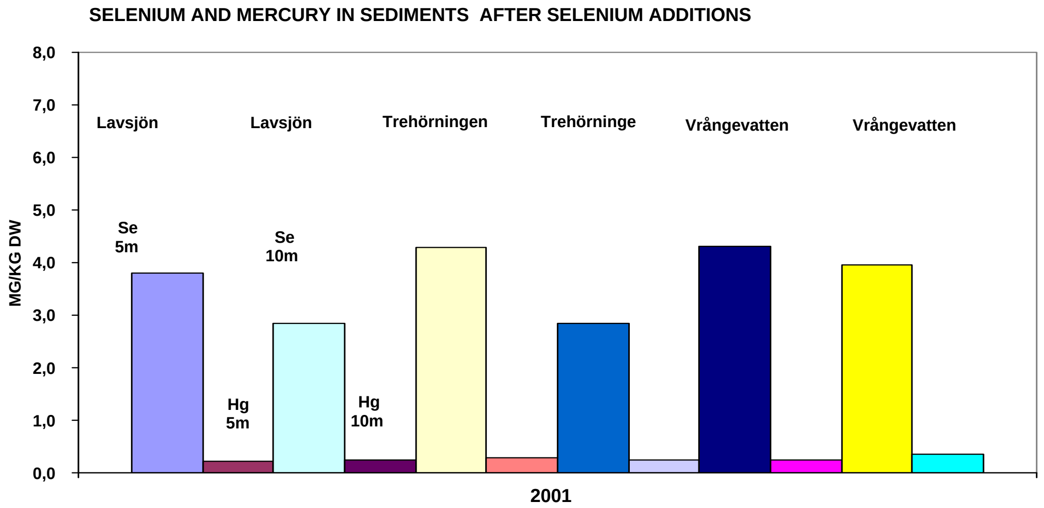

Figure 7

3.4 Effects on plants and invertebrates

Figure 8 below show selenium concentrations in the aquatic moss *Fontinalis* taken from the outlet stream of Lake Lavsjön. It can be seen that the selenium concentrations increase when selenium is added to the water. When the selenium concentration in the water falls during 2001, the concentration in the aquatic moss also falls quickly because the treatment stopped during the previous year. The content of total mercury in the aquatic moss did not decrease in relation to the increase in selenium.

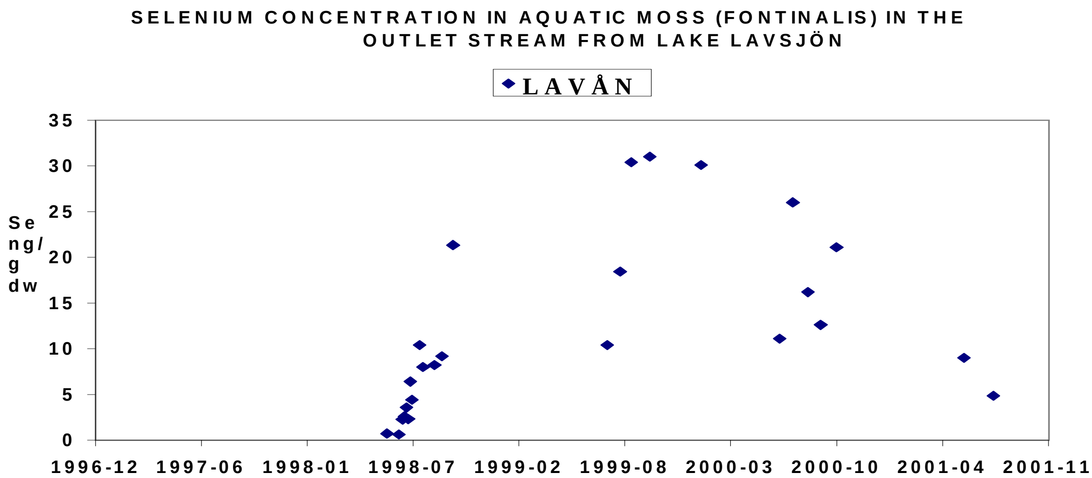

Figure 8

The amounts of selenium increased in all trophic levels as a result of uptake in the food chain in the lakes. Figure 9 below shows that the increase in selenium resulted in a reduction in the methyl mercury concentrations in zooplankton. The concentrations are given in dry weight (DW) which is around 5% of the wet weight. In the wet weight the concentrations are around 20 times lower.

A reduction in the methyl mercury concentrations in the lowest trophic level in the lakes means that all the other trophic levels will be affected. All the higher levels in the food chain will have reduced concentrations of methyl mercury, because a smaller quantity of methyl mercury is available lower down the chain.

This is perhaps one of the most important results of the experiments involving the addition of selenium to the three lakes.

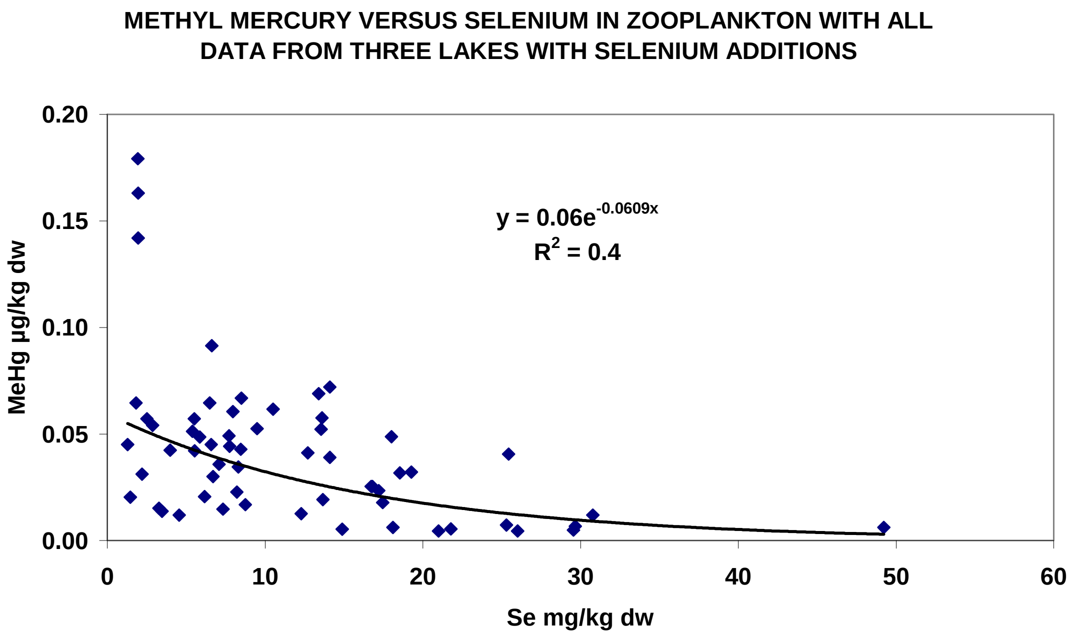

Figure 9

The species composition of zooplankton in the three lakes is otherwise normal for woodland lakes (Lake Lavsjön) and for lakes after many years of liming (Lakes Trehörningen and Vrångevatten). Frozen bag net samples of zooplankton were characterised by species and any developments as a result of the treatment with selenium. Downstream of Lake Lavsjön, the upper Skinnmuddselet was also affected by the addition of selenium, but as only around 15% of the upper Skinnmuddselet's catchment is from Lake Lavsjön, there was a significant dilution of the selenium concentrations in the reservoir.

The species of zooplankton were identified in the frozen samples, which was very time-consuming. As a result some of the smaller species, or those which occurred infrequently, may have been overlooked. However, the characterisation process does allow larger changes to be monitored and also shows how "pure" the samples are.

As shown in Table 2 below, no dramatic or permanent changes could be identified in the mutual abundance relationships of the zooplankton in the lakes from which the samples were taken. Instead, the samples show that the normal variations through the year continued to a large extent, with a certain degree of variation between the years, which was probably caused by variations in both climatic and hydrochemical factors.

In the majority of the lakes and in different parts of the Skinnmuddselet reservoir, this means a preponderance of cladocerans (*Daphnia* and *Bosmina*) during a large part of the growing season. This alternated during the winter with a predominance of copepods. The occurrence of *Holopedium* meant that the work of concentrating the zooplankton samples was very time-consuming because of the water-retention properties of the slime. However, in Lake Vrångevatten in Bohuslän, the calanoid copepod *Eudiaptomus gracilis* makes up a large proportion of the zooplankton during large parts of the year. This is typical of many woodland lakes in Southwest Sweden.

On the basis of the zooplankton samples it can be concluded that the dosages of selenium used in these treatments have not had any significant effect on the zooplankton in the lake systems.

Table 2. Species composition and relative quantities of zooplankton

Hg-Selen Karakterisering av hŒvprover med avseende pŒ zooplankton och ev. utveckling

Färg=selentillförsel

Lake	Date	Daphnia	Bosmina	Holopedium	Chydoridae	Bythotrephes	Leptodora	Polyphemidae	Eudiaptomus	Cyclopoida	Calanoida	Kellicottia	Keratella	Polyarthra
LAVSJÖN	01-10-01	+++	(+)							(+)	(+)			
	02-06-04	+	+							++++	+	(+)	(+)	
	02-06-12	++	++							++	(+)	+	(+)	
	02-07-01	++	++							++	(+)	(+)		
	02-07-05	++	+++							(+)	(+)	(+)		
	02-07-09	+++	+++							+	(+)	(+)		
	02-07-13	++++	++							+	(+)	(+)		
	02-07-16	++++	+							+	(+)			
	02-07-20	++++	+							+	(+)	(+)		
	02-07-23	++++	+							+	+		(+)	
	02-07-30	+++++	+							+		(+)		
	02-08-06	+++	+							+		(+)		
	02-08-11	+++	+							(+)	(+)	(+)		
	02-08-20	+++++	+				?	(+)		+	(+)	(+)	(+)	
	02-08-27	+++++	+							+		(+)		
	02-09-04	+++	++		(+)					+	(+)		(+)	
	02-09-10	++++	+							(+)	(+)	(+)	(+)	
	02-09-18	+++	(+)							(+)	(+)		(+)	
	02-09-25	+++	+							(+)			(+)	
	02-10-01	+++++	(+)							+	(+)		(+)	
	02-10-09	++++	+		(+)					(+)	(+)		(+)	
	03-06-22	++	+++							+	(+)	(+)		
	03-07-07	++	+++							(+)		(+)		
	03-07-27	++++	+							(+)				
	03-08-02	++++	(+)					(+)		(+)				
	03-08-11	+++	+							+	(+)	(+)		
	03-08-27	+++	++				?			+	(+)	(+)		
	03-09-07	++++	+							+	(+)		(+)	
	03-09-26	++++	+							(+)				
	03-10-15	+++	+							++	(+)			
	04-05-29	+	+			(+)				+++	+	(+)		
	04-05-30	+	+							+++	+			
	04-06-15	(+)	+++							+		(+)		
	04-07-09	++	+							++	(+)	+		
	04-08-03	+	+							+	(+)	(+)		
	04-08-31	+++	++							+	(+)			
	04-09-30	+	(+)							(+)	(+)			
ÖVRE SKINNMUDDSELET	01-10-01									(+)			(+)	
	02-06-09	+	+				(+)			+				
	02-06-11			?		(+)				+++++				
	02-07-15	++++	+			(+)	(+)			+	(+)	(+)	(+)	
	02-08-26	(+)	(+)											
	02-10-13	++++	(+)							(+)	(+)		+	
	03-06-22	+	+++							+			(+)	
	03-08-22	++	(+)				(+)			+				
	03-09-03	++++	(+)							+				
	03-10-05	++++	+							(+)				
NEDRE SKINNMUDDSELET	02-07-15	+++	(+)							+		(+)		
	02-07-29	+++	(+)							(+)		+		
	02-08-24	++								++				
	02-10-13	+++	(+)							+	(+)	(+)		
	03-07-16	++++	+							(+)		(+)		
	03-08-02	++												
	03-09-03	++	++							(+)		(+)		
	03-10-05	++	++							(+)				
STOR-TÄLL-VATTNET	01-10-02	(+)	(+)							(+)			(+)	(+)
	02-06-07	(+)	(+)							(+)		(+)		
	03-10-06	++	++							+	(+)	(+)	(+)	
GÄDD-SJÖ-DAMMEN	04-08-09	++	+							+	(+)			
	04-10-02	(+)	(+)							+		(+)	(+)	
	04-11-08	(+)	(+)							+++	(+)	(+)		
VRÅNGEVATTEN	02-10-22	+	+++			(+)			+	+		(+)		
	03-07-19	+	++++						++	(+)		(+)		
	03-08-05	+	++			(+)			++	(+)		++		
	03-08-28	(+)	+++						++	+				
	03-09-17		+++						++	(+)				
	03-11-18	(+)	+++						+++	(+)				
	04-05-11	(+)	+	+					+	(+)				
	04-05-25		+	+		(+)	(+)		+	+				
	04-06-19	+	+	(+)		(+)			++					
	04-07-26	(+)	+	(+)		(+)			++	+		++		
	04-08-27	+	+						++	(+)		(+)		
	04-10-08	+++	+						+	+		(+)		
TREHÖRNINGEN	03-07-17		++							++		(+)	(+)	
	03-07-21	(+)	+							++		++	+	
	03-08-07		(+)							(+)		+++	+	(+)
	03-08-27	(+)	++							(+)		+		
	03-09-21	+	+							+		(+)	+	
	04-05-25	+	++							(+)		++		
	04-05-26	+	+						(+)	++		++	(+)	
	04-06-17	(+)	++							(+)		++	+	
	04-07-30	(+)	++++				(+)			(+)		(+)		
	04-08-29	(+)	++++							(+)				
	04-10-06	(+)	++							(+)		++	+	

\+ = relativ abundans i proven
(+) = enstaka exemplar
? = ej säker identifiering

The selenium concentration has increased and the mercury concentration has decreased in both 8-10 cm long crayfish and in 10-12 cm long crayfish in Lake Vrångevatten (Figure 10). The selenium concentration has increased most and the mercury decreased most in the smaller crayfish.

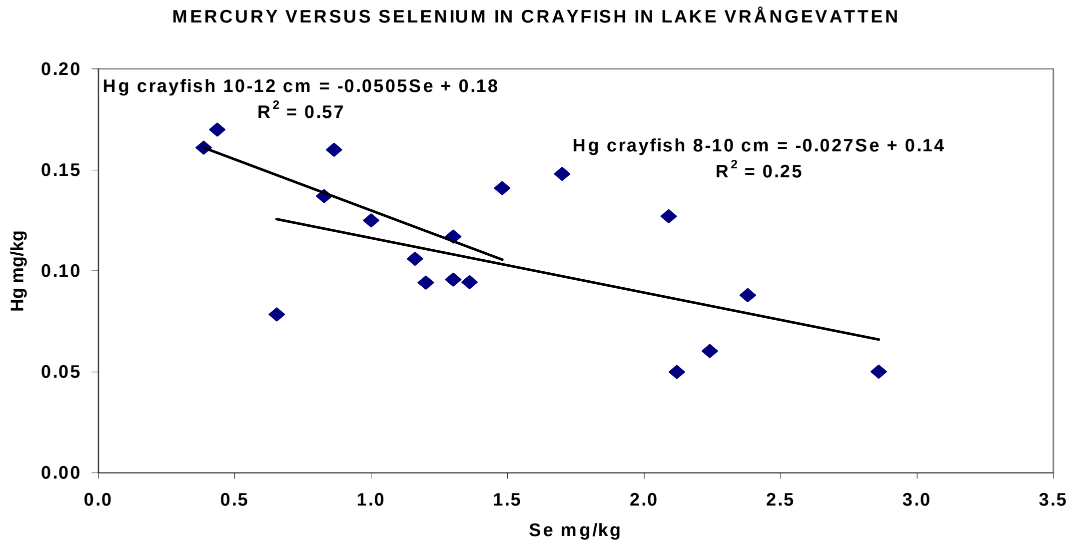

Figure 10

The selenium concentrations increased in all the different types of insect larvae examined after the selenium dosages were added to the lakes.

Figure 11 shows the increase in the selenium concentration in predatory insect larvae, in this case, large dragonfly larvae from the three lakes. During 2001 the selenium concentrations fell.

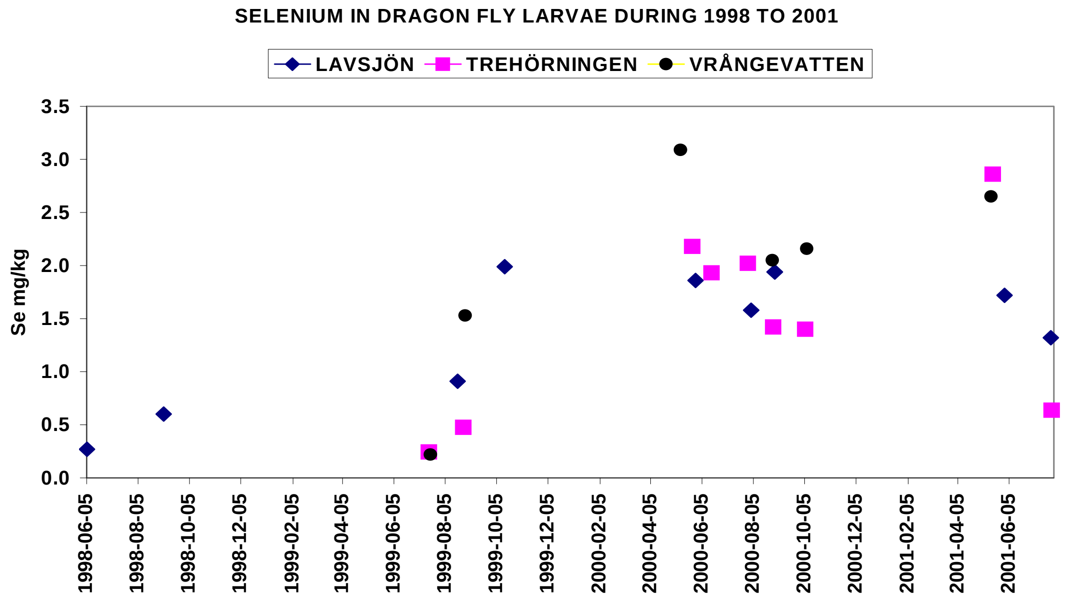

Figure 11

Figures 12 to 17 show that the mercury concentrations in large and small dragonflies, damselflies and caddis flies have not been affected by the high selenium concentrations in any of the three lakes.

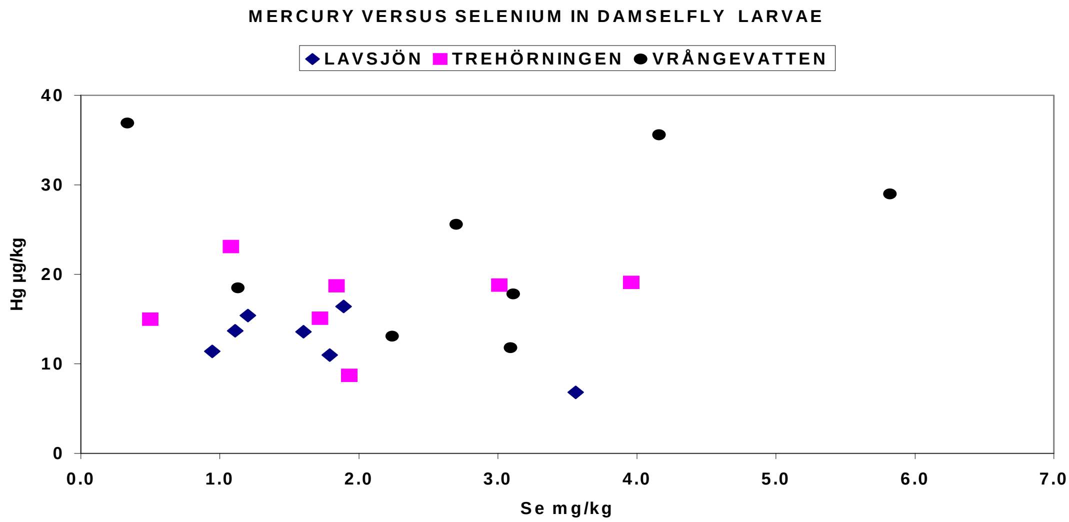

Figure 12

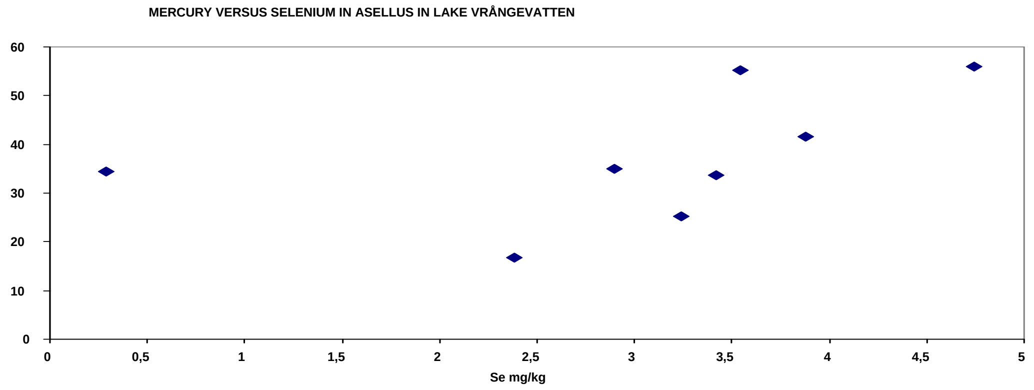

Figure 13

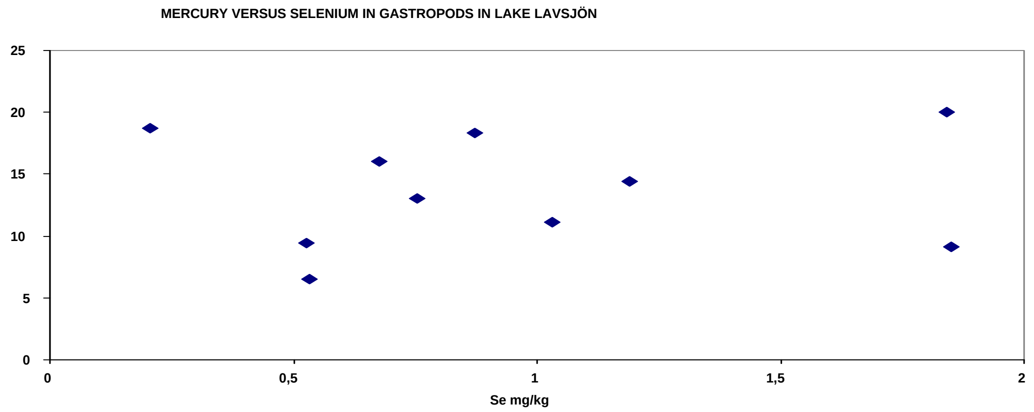

Figure 14

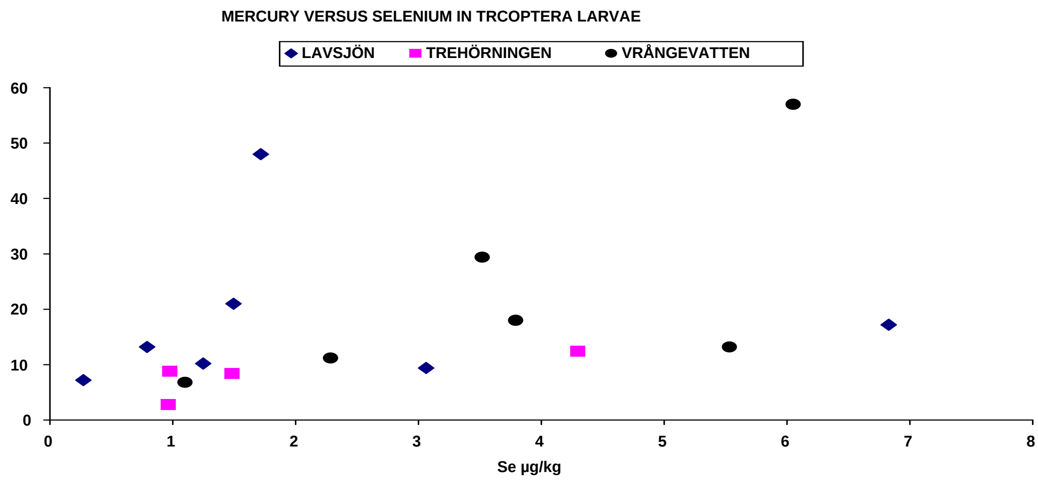

Figure 15

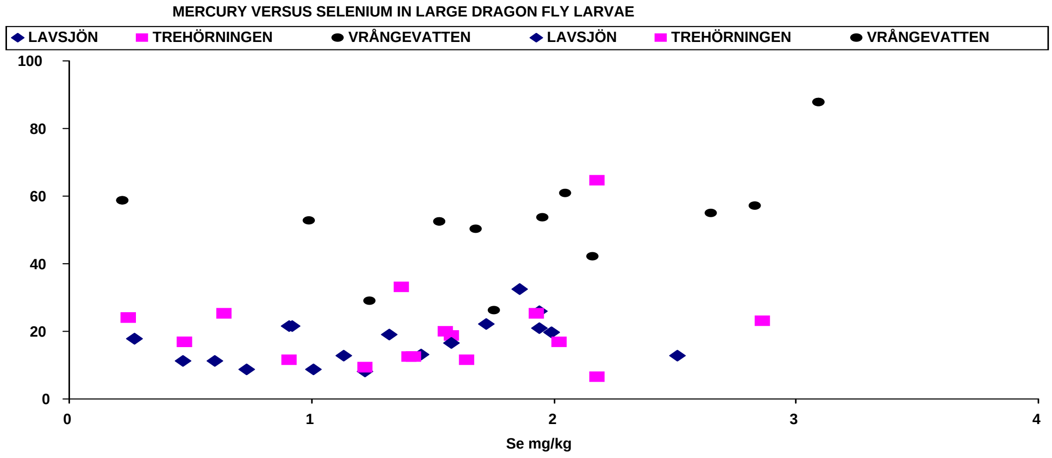

Figure 16

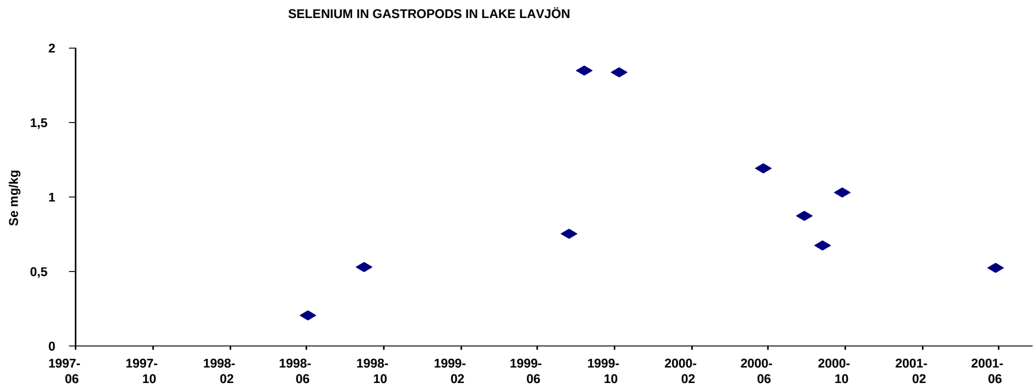

Figure 17

3.5 Effects on fish

The samples taken by netting fry and fish with a fine-mesh net have not shown any negative effects on the reproduction of roach and perch. During 2001 a large number of 1 and 2 year old perch were caught, which were born during the years when Lake Lavsjön had selenium added to its water.

Similarly, roach fry were caught every year during the summer months in all three lakes. Figure 18 below shows that the concentration of methyl mercury varied significantly with low concentrations of selenium in roach fry. Some fry had very high concentrations of methyl mercury. After selenium was added to the water, the concentration of selenium in roach fry increased and the concentration of methyl mercury decreased. The addition of selenium resulted in a 50% reduction in methyl mercury in roach fry in the three lakes.

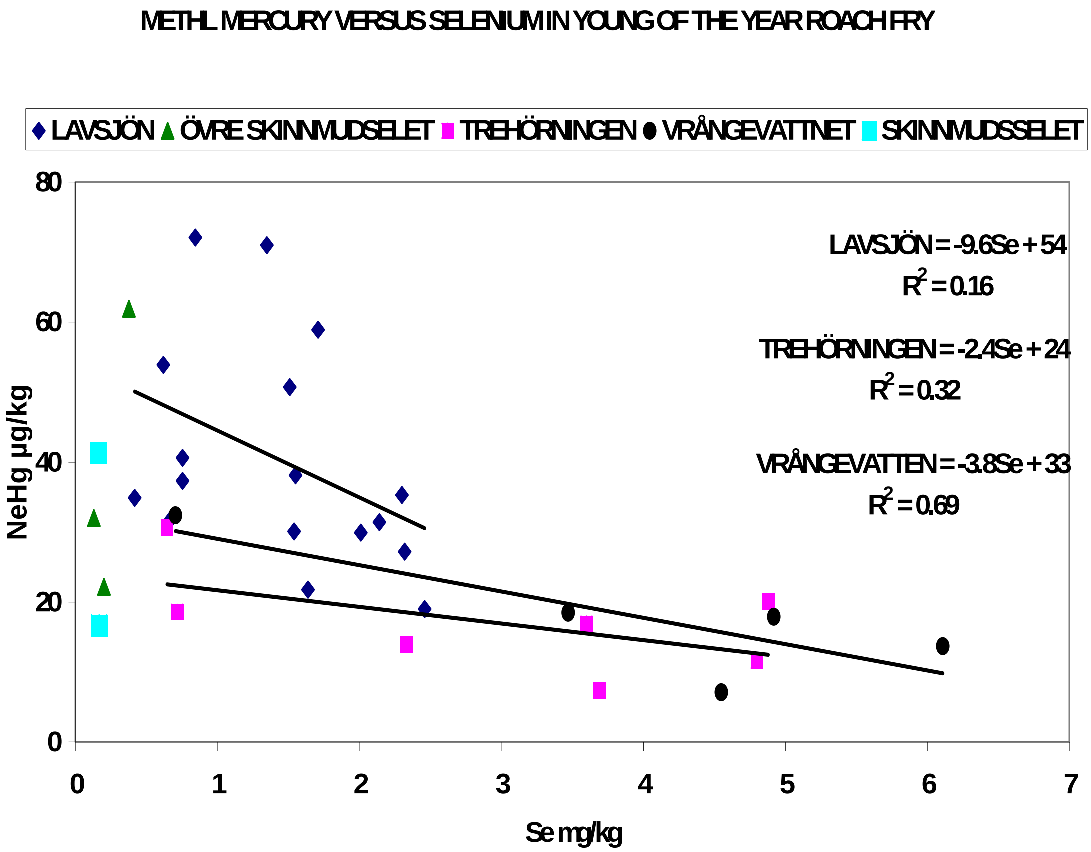

Figure 18

The selenium concentrations in young roach increased in the three lakes, which resulted in reduced concentrations of methyl mercury in the muscles of the fish (see Figure 19 below).

METHYL MERCURY VERSUS SELENIUM IN 4-6 cm ROACH

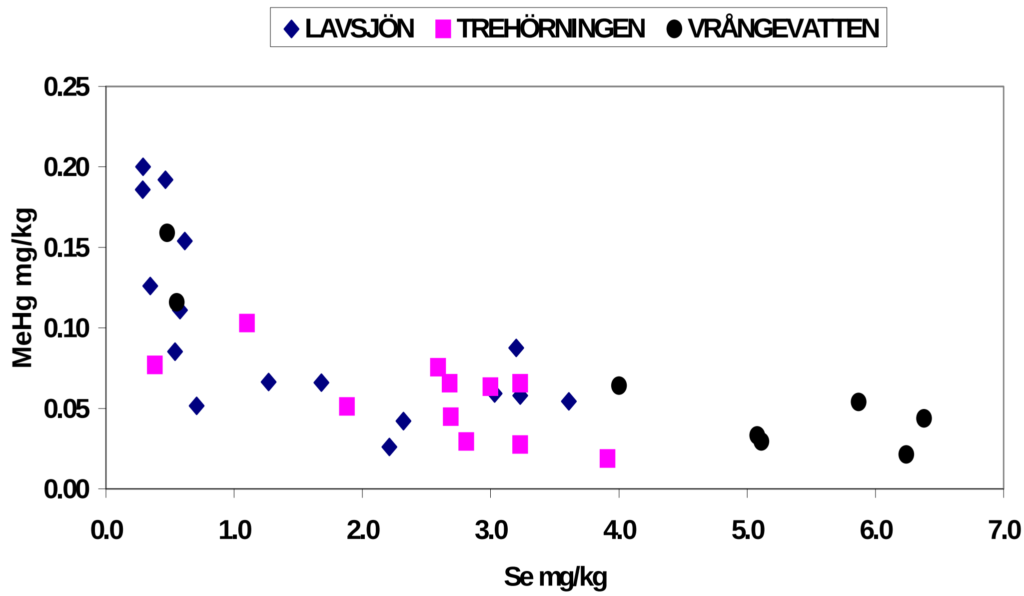

Figure 19

METHLM MERCURY VERSUS SELENIUM IN 12 TO 14 CM ROACH IN LAK LAVSJÖN, LAKE TREHÖRNINGEN AND LAKE VRÅNGEVATTEN

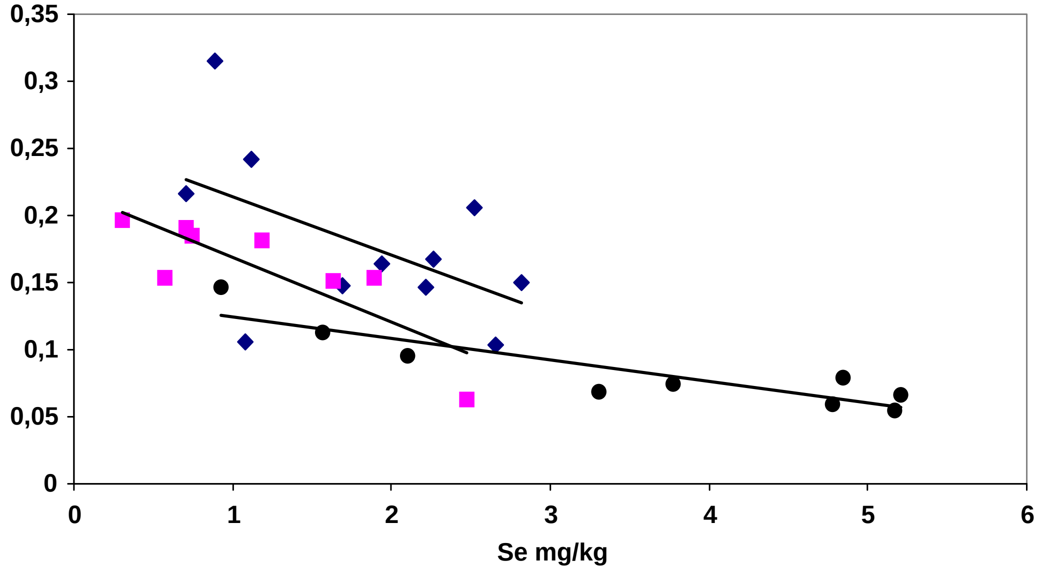

Figure 20

In the same way as in roach, the selenium concentration in younger perch has increased, and the methyl mercury concentration decreased, as shown in Figures 21 to 23 below. The rapid increase in selenium and the corresponding reduction in methyl mercury in younger perch and roach are a result of the fact that this size of fish feeds primarily on zooplankton. Figure 8 above showed that zooplankton responded quickly to the addition of selenium, with an increase in selenium concentrations and a reduction in methyl mercury concentrations, and this was reflected quickly higher up in the food chain.

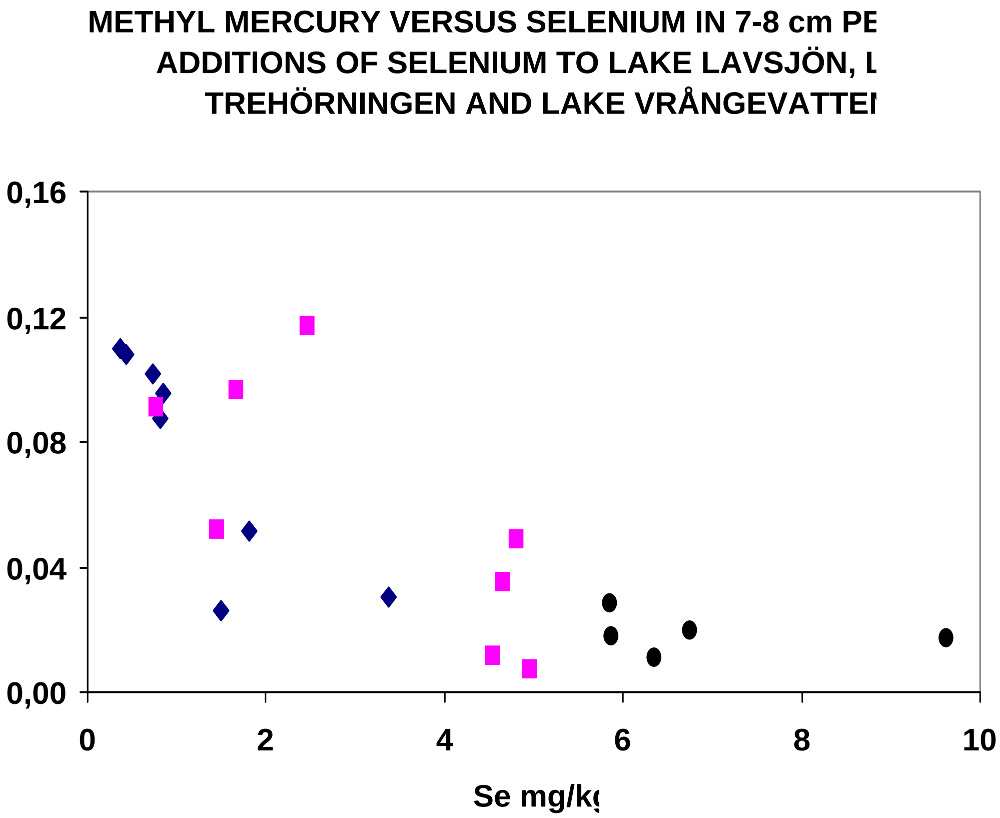

Figure 21

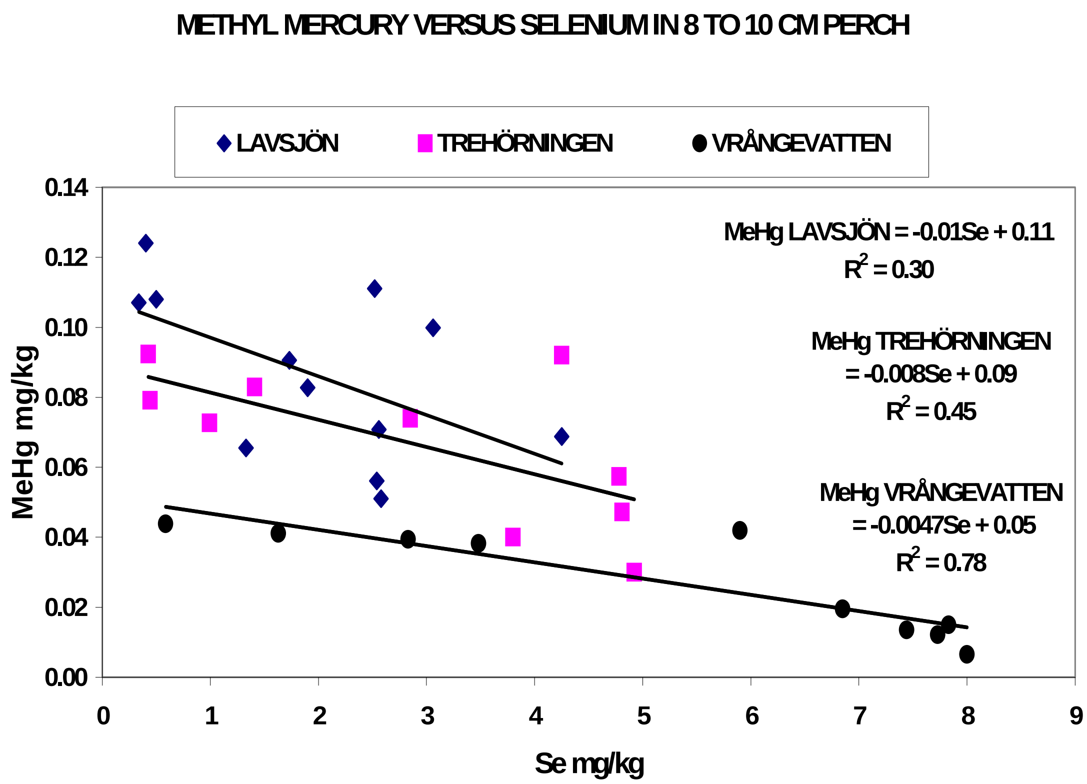

Figure 22

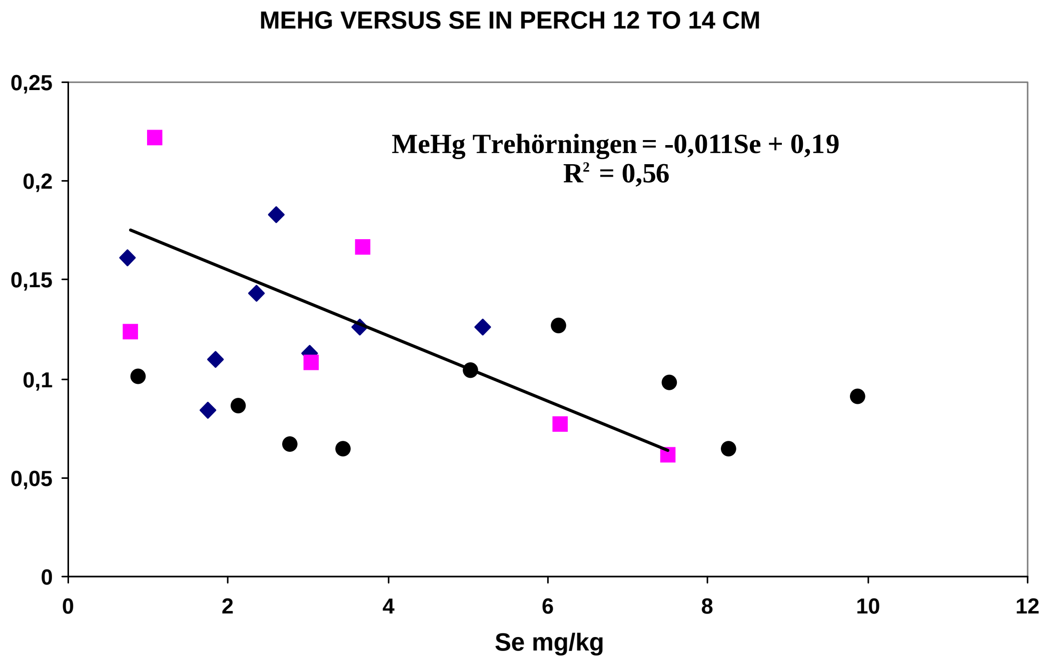

Figure 23

Northern pike show a major reduction in methyl mercury concentrations in the weight groups 0.4 to 0.8 kg (Figure 24) and 0.8 to 1.2 kg (Figure 25). The reduction in methyl mercury takes place at the same time as the selenium concentrations in the muscles are increasing. There is a 50% reduction in methyl mercury in pike from both Lake Lavsjön and Lake Vrångevatten. In Lake Vrångevatten, which has the largest increase in selenium, there is the greatest reduction in methyl mercury. Pike in these size groups, which showed such a rapid decrease in methyl mercury, feed on young roach and perch, which showed the greatest reduction in methyl mercury (see Figures 19 to 23).

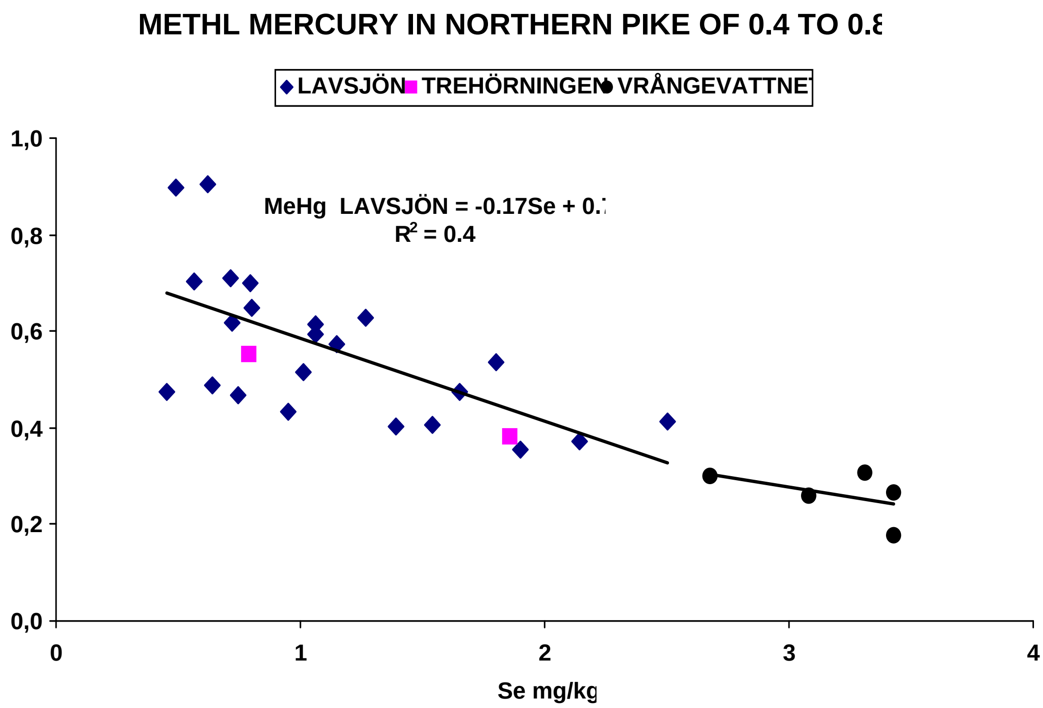

Figure 24

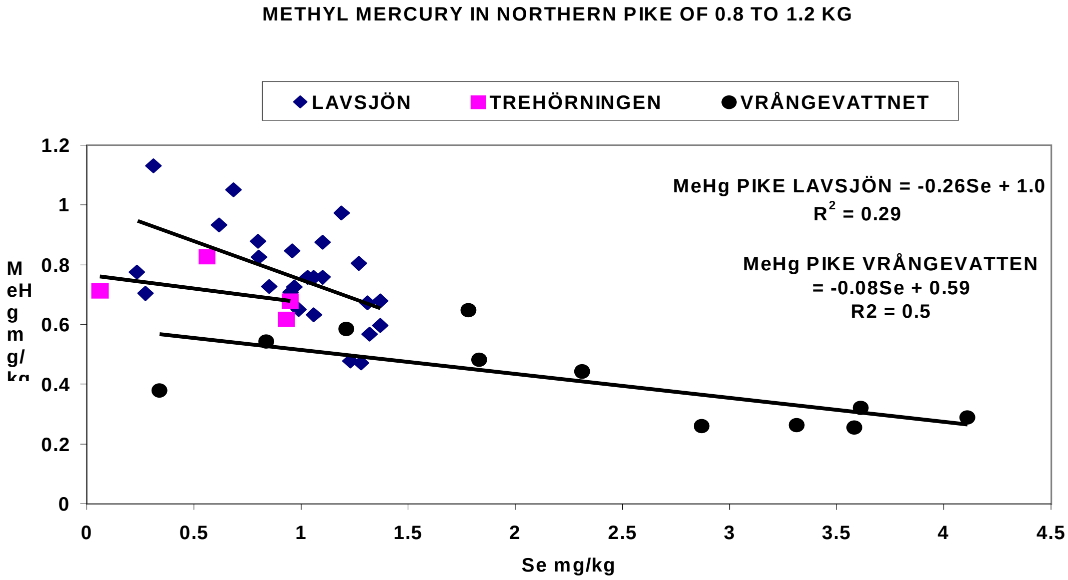

Figure 25

Older pike (Figure 26) and perch show increased selenium concentrations, but in the older pike this has not yet led to a significant reduction in methyl mercury concentrations. Large pike eat mainly older and larger prey fish, which have not shown the same reductions in methyl mercury as younger roach and perch. There is a delay of a few or of several years in the effect on the older pike. Only when the roach and perch with reduced concentrations grow older and larger do they become the prey of large old pike.

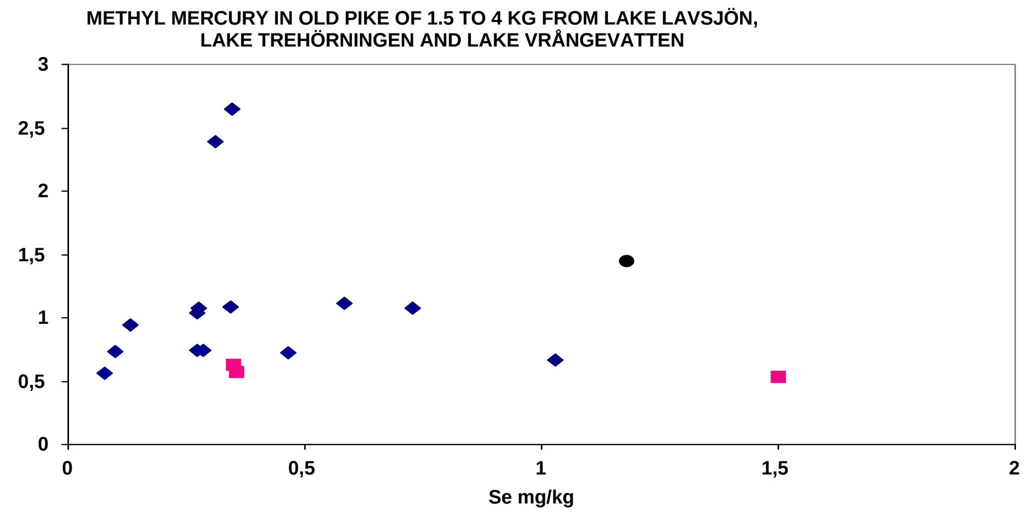

Figure 26

Figures 27 and 28 below show that even larger (20-22 cm) roach and perch have lower concentrations of methyl mercury corresponding to the increase in selenium concentrations in the three lakes.

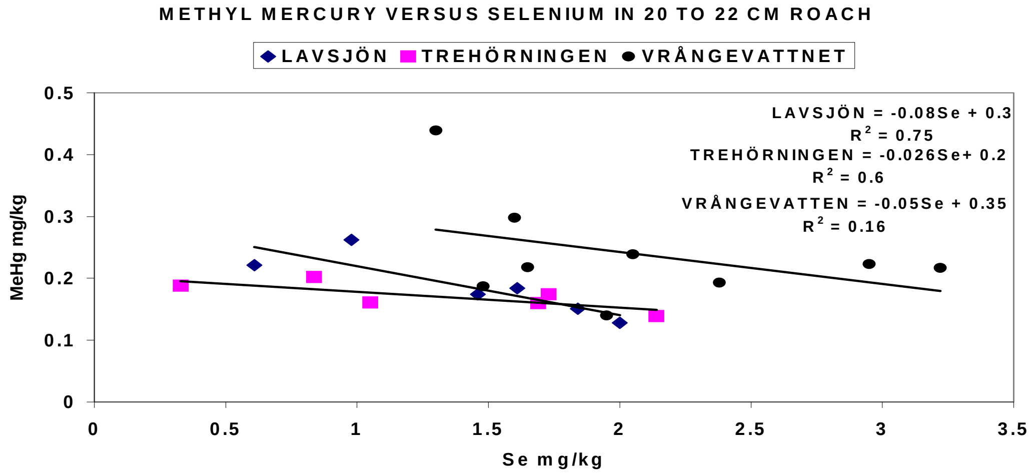

Figure 27

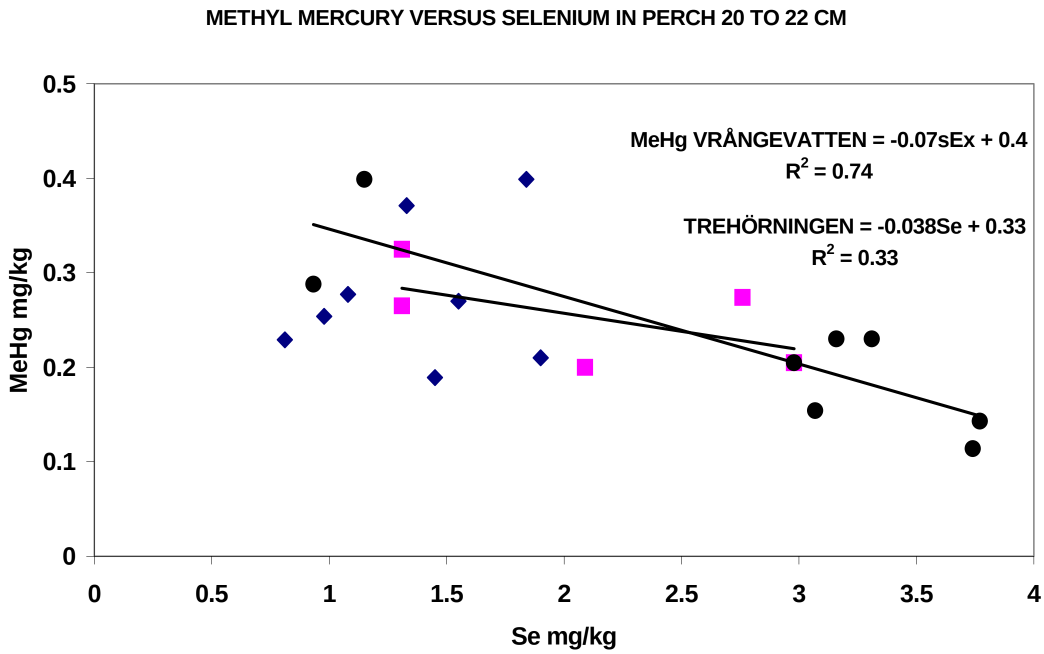

Figure 28

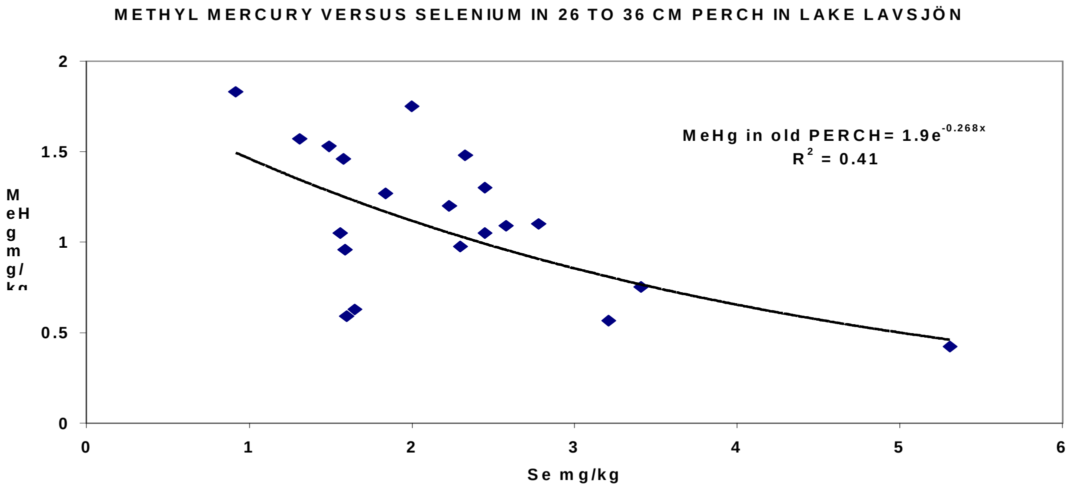

Figure 29

Figure 29 below shows that older perch (26 – 36 cm) from Lake Lavsjön have reduced methyl mercury concentrations and increased selenium concentrations in their muscles. Not enough older perch from Lake Trehörningen and Lake Vrångevatten were examined to be able to draw a connection between the increased selenium and reduced methyl mercury concentrations.

Figure 30 shows that bream have low methyl mercury concentrations, which reduce as the concentrations of selenium in the muscle increase. At a selenium concentration of 2 mg/kg in the muscles, the methyl mercury concentrations have reduced by around 60%.

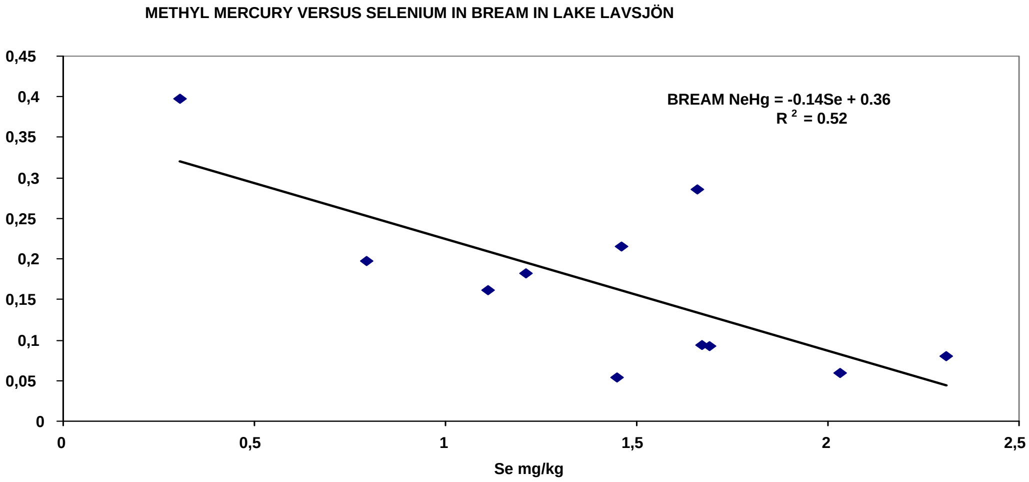

Figure 30

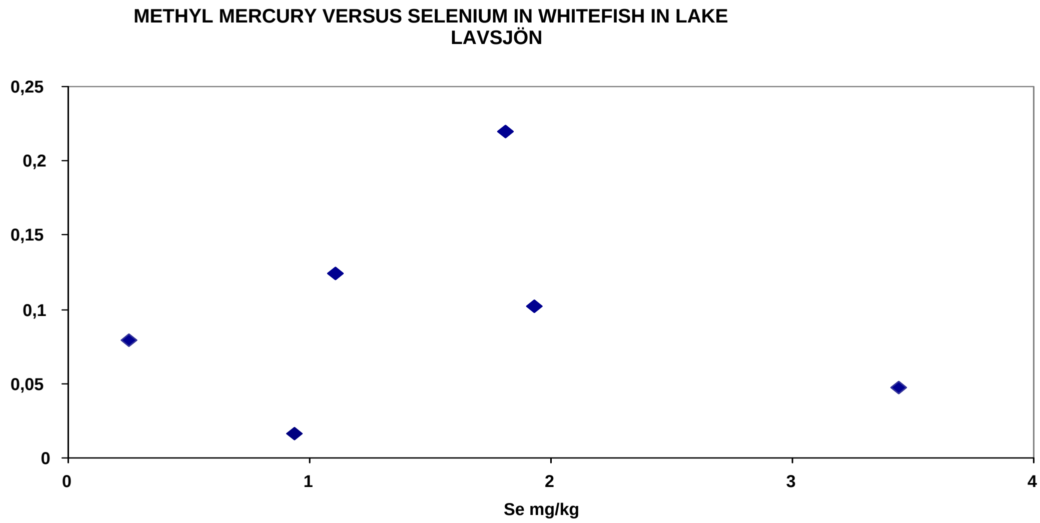

Figure 31

Whitefish in Lake Lavsjön also have low concentrations of methyl mercury and there is no connection between selenium and methyl mercury in the fish muscles as shown in Figure 31. Individual analyses of small burbot and bleak show that selenium is absorbed and increases in concentration in the muscles, but there is not enough data to be able to draw a connection with methyl mercury in these species.

4 Effects of selenium in the Skinnmuddselet reservoir

In summer the Skinnmuddselet reservoir has an area of 27 km^2 and a volume of 150 million m^3. It lies immediately downstream of Lake Lavsjön, which was treated with selenium. The top part of the reservoir is called Lanaträsket and this is where the river Lavån enters the reservoir. The flow of water in the Lavån from Lake Lavsjön to the reservoir makes up around 15% of the reservoir's annual catchment. Downstream of Lanaträsk, the reservoir becomes the upper Skinnmuddselet, which stretches as far as road 92. The lower part of the Skinnmuddselet reservoir is the part between the road 92 and the outflow into the Stennäs power station. After 1999, the uptake of floating peat moved from Stennäs to the Gäddsjödammen, which is a large bay in the lower part of the Skinnmuddselet. The Skinnmuddselet drains through the river Gideälven and the lake immediately downstream of the reservoir, which is called the Stora Tällvattnet.

Below is a rough calculation of the selenium concentrations in different parts of the reservoir. Based on information about the daily flow of water into the reservoir during July, August and September, the average daily flow from Lake Lavsjön to the Skinnmuddselet was calculated at 4.2, 1.5 and 5.2 m^3/s respectively during 1998, 1999 and 2000. The amount of water, which flowed through during the three summer months, was 34, 12 and 41 million m^3 during the three years when the lake was treated. The selenium concentration reduced to a background level several times during the first year in Lake Lavsjön, which has a volume of 2 million m^3. This resulted in higher dosages being used in 1998. The quantity of selenium used was around 18, 10 and 15 kg respectively in each of the three years. In theory this should result in a calculated average selenium concentration in Lake Lavsjön of around 0.4, 0.5 and 0.4 µg/l for the respective years. The calculations give a slightly higher concentration than the measurements during the first and second year and a slightly lower concentration during the third year. Analyses of the water showed that Lake Lavsjön had an average selenium concentration of around 0.05 µg/l during the summer months before the treatments. The selenium concentration increased to around 0.3, 0.4 and 0.5 µg/l during the years when selenium was added. The absorption of selenium in the sediment and other uncertain factors may explain the differences between the measured and calculated concentrations.

The quantity of water with an increased selenium concentration flowing into the Skinnmuddselet reservoir is greatest during the first and third years, when around 30 to 40 million m^3 flowed into the reservoir from Lake Lavsjön. This should be compared with the total volume of the reservoir, which is around 150 million m^3. The extremely high levels of precipitation in 1998 and 2000 also resulted in the flow from other parts of the reservoir's catchment area being equally high. This led to a significant dilution of the selenium concentration in the reservoir. The dilution was estimated to be around one-third in the upper Skinnmuddselet and in the part of the Skinnmuddselet closest to road 92. In the lower part of the Skinnmuddselet and the Stora Tällvattnet the dilution of the selenium content compared to Lake Lavsjön was estimated at one fifth. The calculated selenium concentration in the water of the upper part of the reservoir was 0.1 and 0.2 µg/l during 1998 and 2000 respectively. The lower part of the reservoir and the Stora Tällvattnet had selenium concentrations of 0.06 and 0.1 µg/l.

Figure 32 shows the average concentrations of selenium from individual water analyses in the upper Skinnmuddselet, Skinnmuddselet and in the Stora Tällvattnet before and after the selenium treatment, together with the average concentrations in Lake Lavsjön. The selenium concentrations have increased in both parts of the reservoir and in the Stora Tällvattnet from around 0.03 µg/l to around or above 0.10 µg/l. The flow to the Lanaträsket, which is directly downstream of Lake Lavsjön, consists entirely of the river Lavån, and the water chemistry in this part of the reservoir is the same as that in Lake Lavsjön. The average concentrations of selenium in the Lanaträsket can therefore be assumed to have been the same as in Lake Lavsjön.

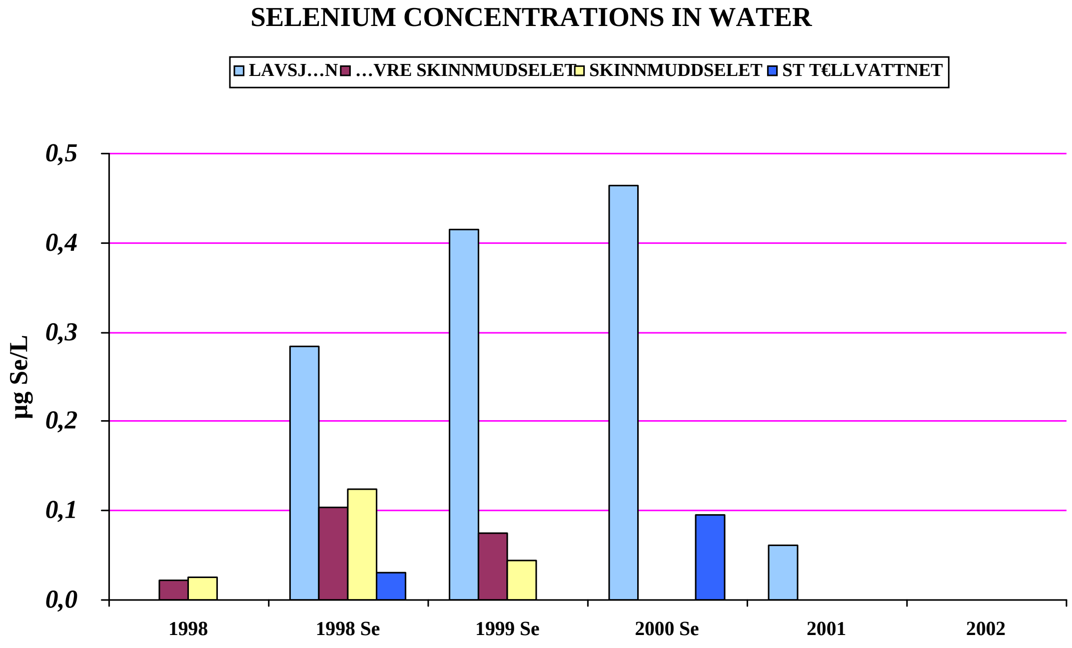

Figure 32

Individual analyses of selenium in zooplankton in the upper and lower parts of the Skinnmuddselet also show an increase related to the addition of selenium to Lake Lavsjön (Figure 33). Selenium in zooplankton increased three- to fourfold in the upper Skinnmuddselet during 1998, which corresponds to the increase in selenium in the water.

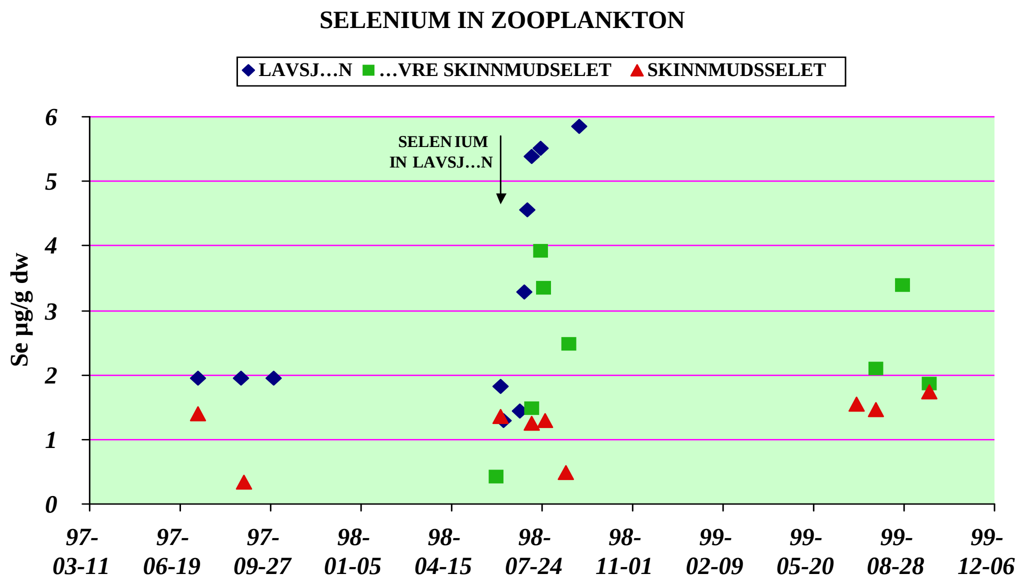

Figure 33

A few analyses of selenium in young perch (10 cm) are available. These show that an increase in selenium concentrations in the water and the zooplankton resulted in an increase in selenium in young perch. The largest increase in selenium in young perch was found in the Lanaträsket, but there is also a clear increase in the upper parts of the Skinnmuddselet.

Figure 34 shows that concentrations of methyl mercury in fish muscles reduced significantly in young perch. The reduction took place in all the areas of the Skinnmuddselet, which were examined and in the Stora Tällvattnet during 1999, 2000 and 2001 after the addition of selenium to Lake Lavsjön.

The two factors, which could explain this, are:

- the increased selenium concentrations in the fish muscles

And

- a reduction in the exposure to methyl mercury in the water because of the change in the treatment of floating peat in the reservoir.

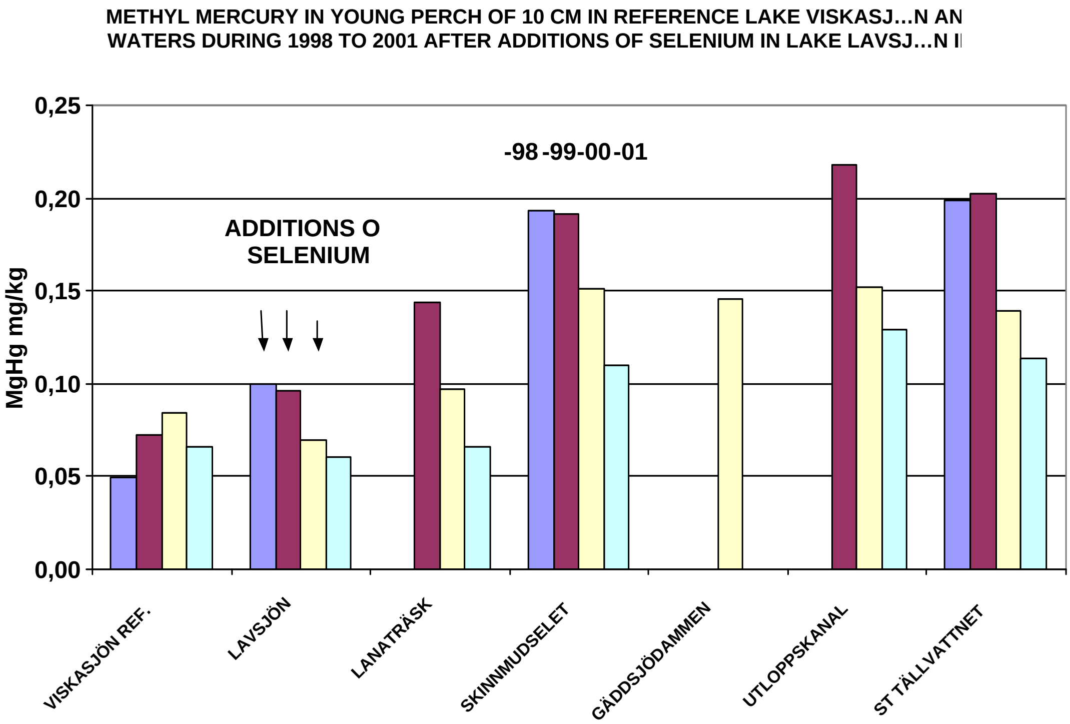

Figure 34

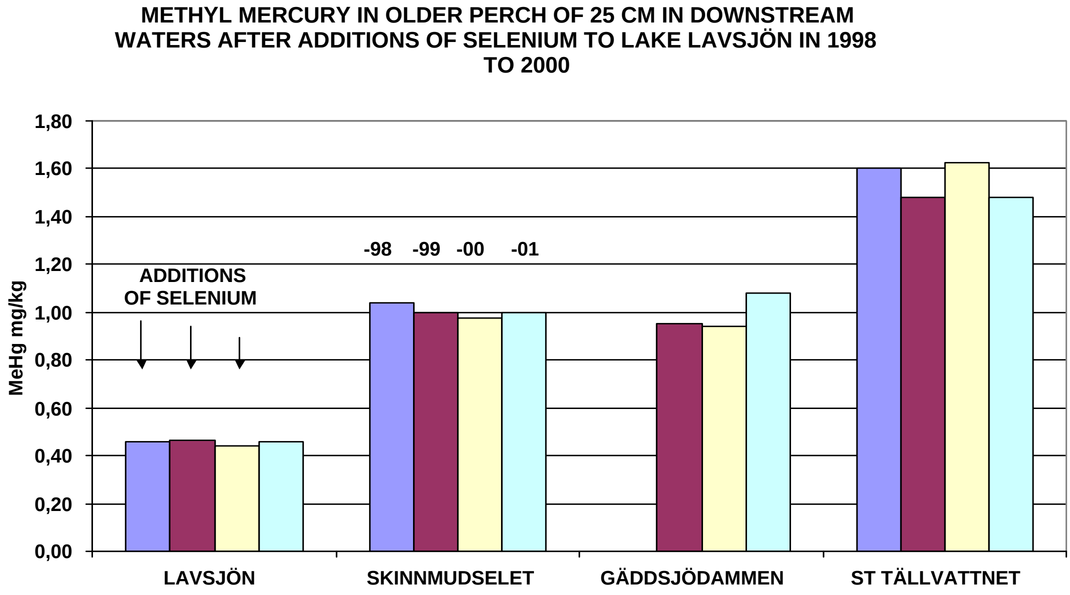

Figure 35

Figure 35 shows that the methyl mercury in older perch in different parts of the Skinnmuddselet and the Stora Tällvattnet did not reduce during the years of selenium treatment in Lake Lavsjön. Follow-up studies of selenium in fish are needed. This data is important to allow a quantification of the significance of selenium for the reduction of methyl mercury in fish at a selenium concentration in water of 0.1 – 0.2 µg/l.

5 Conclusions

Selenium was added to Lake Lavsjön in Västerbotten in 1998, 1999 and 2000. Selenium was added to Lake Trehörningen in the county of Örebro and Lake Vrångevatten in Västra Götaland in 1999 and 2000. A low dosage of selenium was used which corresponds to an average five- to sevenfold increase in the lakes' background concentrations. The average concentration of selenium in the lake water varied between 0.29 and 0.53 µg/l during the years when selenium was added to the three lakes. The concentration of selenium in the sediment in the three lakes showed a twofold increase. The selenium was added in the propeller wake of a boat using a solution of sodium selenite with selenium content of 45.2%. The selenium treatment in Lake Lavsjön was very labour-intensive, with one or more treatments per month during the period from June to October. This is caused by the lake's naturally short water retention period and by the high levels of precipitation in the years of the trial. Only three selenium treatments were carried out during each of the two years of the trial (1999 and 2000) in Lakes Trehörningen and Vrångevatten.

The increased concentrations of selenium in the water resulted in a quick uptake of selenium in aquatic moss, zooplankton, several types of insect larvae, roach fry and the youngest age groups of roach and perch. The increase in selenium in aquatic moss and invertebrates did not result in a reduction in the concentration of total mercury. However the increase in selenium did result in reduced concentrations of mercury in crayfish. One very important result is that the increased selenium concentrations in zooplankton resulted in a reduction in the concentration of methyl mercury in zooplankton. There was a rapid increase in the selenium concentrations in roach fry, younger roach and younger perch, which led to a rapid reduction in the methyl mercury in the fish. The selenium concentrations also increased and the methyl mercury concentrations reduced in older roach (10 – 25 cm) and perch after 2 to 3 years of selenium treatment in the lakes. The reduction in the concentrations of methyl mercury in roach fry, younger roach and younger perch was primarily caused by the reduction in the bio-accumulation of methyl mercury in their food sources, which consist mainly of zooplankton. These had lower concentrations of methyl mercury after the selenium treatments.

After two or three years of selenium treatments in the lakes, the concentrations of methyl mercury had reduced by around 50% in pike in the size groups 0.4 – 0.8 kg and 0.8 – 1.2 kg. The speed of this effect in the higher part of the lakes' food chains is a result of the fact that the main food for pike of this size is young roach and perch. The selenium concentrations in older pike increased slightly, but during the short period of the trial no reduction in the concentrations of methyl mercury was observed. In the muscles of older perch (26 – 36 cm) from Lake Lavsjön there were reductions in the concentrations of methyl mercury and increased concentrations of selenium. No negative effects of the selenium treatments in the lakes on plants and invertebrates were observed. In the majority of the lakes and in different parts of the Skinnmuddselet reservoir, there was a preponderance of cladocerans (*Daphnia* and *Bosmina*) during a large part of the growing season. This alternated during the winter with a predominance of copepods. However, in Lake Vrångevatten in Bohuslän, the calanoid copepod *Eudiaptomus gracilis* makes up a large proportion of the zooplankton during large parts of the year. This is typical of many woodland lakes in Southwest Sweden.

On the basis of the zooplankton samples it can be concluded that the dosages of selenium used in these treatments have not had any significant effect on the zooplankton in the lake systems. The reproduction of invertebrates, crayfish, roach, perch, pike, bream, bleak, whitefish and many other species was unaffected. The flow into the Skinnmuddselet reservoir of water from Lake Lavsjön, which had been treated with selenium, resulted in increased selenium concentrations in the water, zooplankton and young perch in the reservoir. The increase in the food chain occurred at a selenium concentration in the water of 0.1 to 0.2 µg/l. The increase in selenium in the muscles of young perch may have contributed to the major reduction in methyl mercury in the whole of the reservoir and the Stora Tällvattnet. The amounts of methyl mercury in older perch in the reservoir and the Stora Tällvattnet did not decrease.

The figures 36, 37, 38, and 39 below show a very uniform decrease in methyl mercury in fish mussel in nearly all of the investigated size classes of roach, young perch, old perch and Northern pike as a function of increased concentrations of selenium in lake water and fish mussel. The figures include methylmercury and selenium analyses of fish from the three treated lakes from both pre-treatment (1997) and treatment years (1998, 1999, 2000 and 2001).

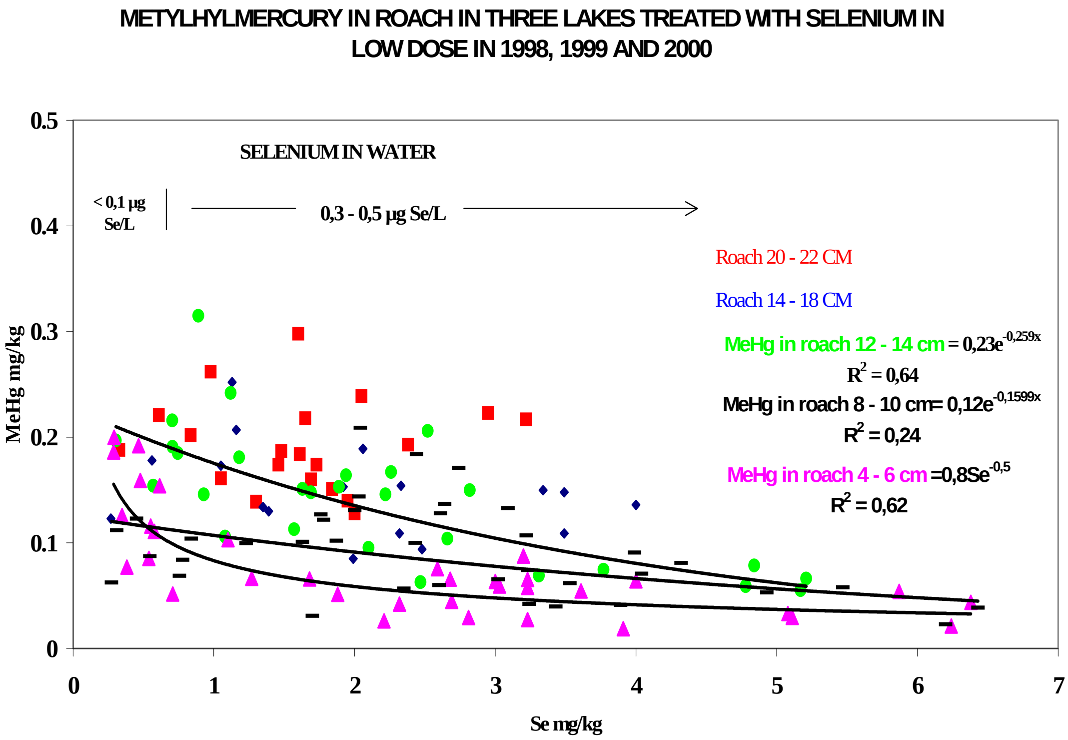

Figure 36

METYLMERCURY I YOUNG PERCH IN THREE LAKES TREATED WITH SELENIUM IN LOW DOSE IN 1998, 1999 AND 2000

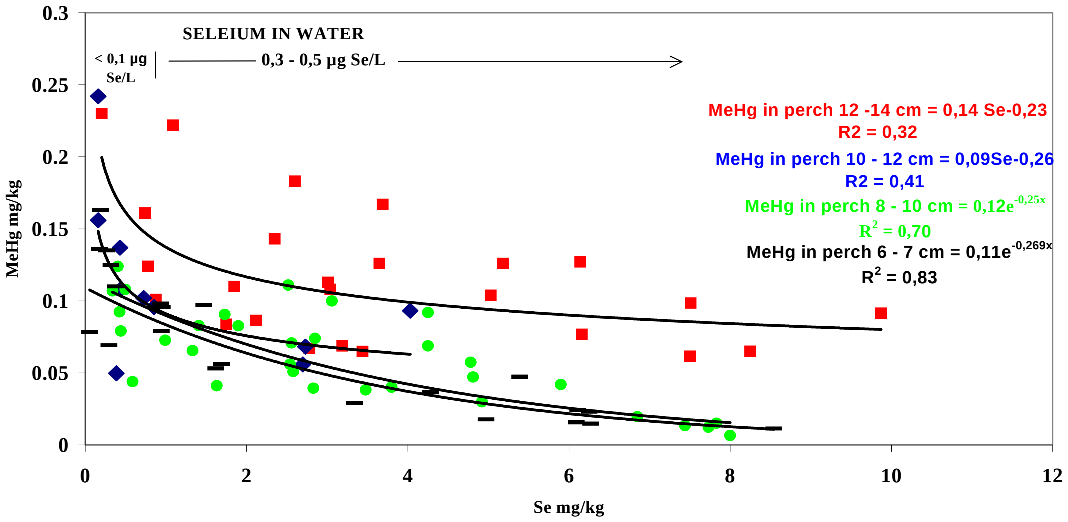

Figure 37

METHYLMERCURY IN OLD PERCH IN THREE LAKES TREATED WITH SELENIUM IN LOW DOSE IN 1998, 1999 AND 2000

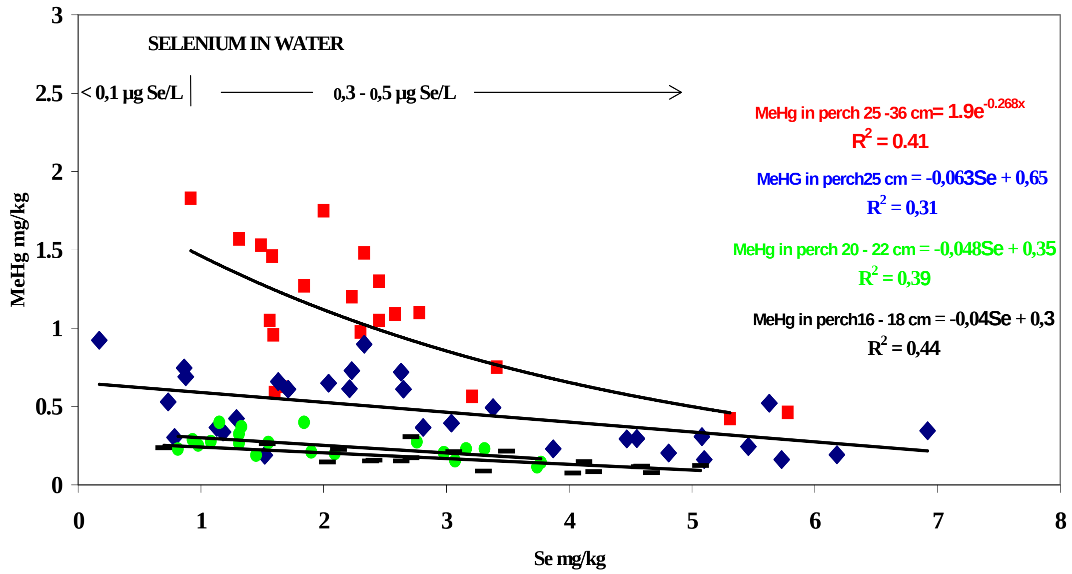

Figure 38

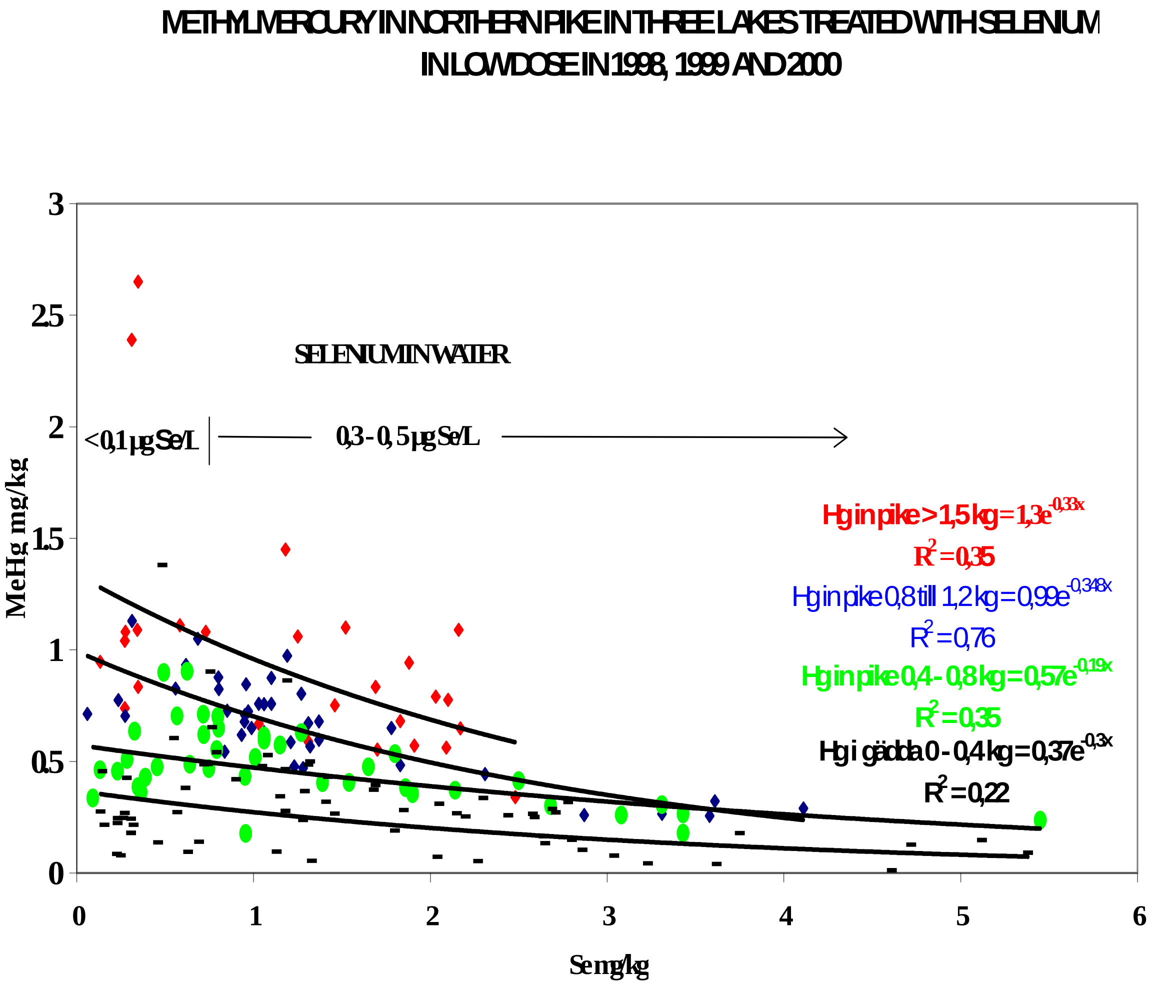

Figure 39

6 Recommendations

- Large-scale trials with a very low dosage of selenium with an average selenium concentration of 0.3 – 0.4 µg/l in the water could start immediately.
- This average concentration of selenium during a period of 2 to 3 years will result in a reduction of around 50% in the methyl mercury in the muscles of younger pike (0.4 – 1.2 kg). At the same time the selenium concentration in the muscles will increase from 0.3 – 0.4 mg/kg to around 2 mg/kg.
- An appropriate large-scale trial can be carried out in the Skinnmuddselet hydroelectric reservoir in the river Gideälven water system in Västerbotten/Västernorrland.

- At the same time it is very important to follow up the ongoing trials in Lakes Lavesjön, Trehörningen and Vrångevatten. The continuation of the studies is necessary to follow up the longer-term effects (> 4 years) of very low dosages of selenium in lakes and reservoirs.
- Follow-up studies of selenium in fish in the upper and lower parts of the Skinnmuddselet and Stora Tällvattnet are needed. This data is important to allow a quantification of the significance of selenium for the reduction of methyl mercury in fish at a selenium concentration in water of 0.1 – 0.2 µg/l.

The project hypothesis has been substantiated by the trials, which have demonstrated that:

The addition of very low dosages of selenium to lakes and storage reservoirs has a twofold positive effect. The methyl mercury concentrations in fish decrease at the same time as the selenium concentrations increase.

7 Literature

Parkman, H. and Hultberg, H., 2002: Occurrence and effects of selenium in the environment – a literature review, IVL publ. B 1486

Fredriksson, R., 2001: Effects on mercury concentrations in zooplankton and of fish of low-dosage treatments of selenium. Examination study at the University of Kalmar.