Agrifood Research Reports 36

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Agrifood Research Finland

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42 p.

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ISBN 951-729-805-6 (Printed version)
ISBN 951-729-806-4 (Electronic version)
ISSN 1458-5073 (Printed version)
ISSN 1458-5081 (Electronic version)
www.mtt.fi/met/pdf/met36.pdf
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MTT Agrifood Research Finland
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Publisher
MTT Agrifood Research Finland, FIN-31600 Jokioinen, Finland
Distribution and sale
MTT Agrifood Research Finland, Data and Information Services
FIN-31600 Jokioinen, Finland
Phone + 358 4188 2327, Fax + 358 4188 2339
e-mail julkaisut@mtt.fi
Published in 2003
Printing house
Data Com Finland Oy
Photographer
Marko Ruusiala and Merja Eurola

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Abstract

In Finland compound fertilizers have been supplemented with selenium (Se) since 1984. In 1998, the Finnish Ministry of Agriculture and Forestry raised the Se supplementation level in fertilizers from 6 to 10 mg Se/kg fertilizer. The reason for this revision was mainly the decreasing trend in the Se content of foods and daily Se intake due, to the lower level of Se supplementation introduced in 1990 and the decreased use of fertilizers.

The change in the Se supplementation level has clearly increased the Se content of feeds, basic foods, and human serum, and the daily Se intake. Since 1998, the mean Se content of wheat and rye has doubled, but the variation in grain samples is large (<0.010-0.260 mg Se/kg dw). The Se content of standard milk has increased about 30% and now fluctuates between 0.20-0.26 mg/kg dw. Also the Se content of meat has increased about 30-50% since 1999. Se concentrations have increased only slightly in eggs and vegetable foods.

The average daily Se intake has increased about 20% since 1999 and is now 0.08 mg/day at an energy level of 10 MJ. In Finland, the average daily Se intake is now higher than in most other European countries and is considered to be a satisfactory and safe level. It satisfactorily meets the RDA and DRI recommendation of an Se intake of 0.055 mg/d. The most important sources of Se are milk, meat, and milk and meat products, which together cover nearly 70% of the total Se intake. After decreasing the amount of fertilizer Se in 1990-1998, serum Se measured in 1999 had decreased to a stable level of 1.10 µmol/l. As a consequence of the second change in the amount of fertilizer Se, mean serum and whole blood Se have started to increase. The serum Se is now at the level of 1.40 µmol/l.

Key words: selenium, fertilizer, fertilization, soil, foods, cereals, milk, meat, feeds, intake, human serum

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Seleenityöryhmän tulokset vuosilta 2000–2001

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Tiivistelmä

Suomessa on lisätty seleeniä moniravinteisiin lannoitteisiin vuodesta 1984 lähtien väestön ja kotieläinten seleeninsaannin turvaamiseksi. Samassa yhteydessä järjestettiin seleenilisäyksen vaikutusten seurata maaperässä, rehuissa, elintarvikkeissa ja ihmisen veren seerumissa sekä seleeninsaannissa. Lannoitteiden seleenimäärää on tänä aikana muutettu kaksi kertaa. Viimeisin muutos oli vuonna 1998, jolloin lannoitteiden seleenipitoisuutta lisättiin maa- ja metsätalousministeriön päätöksellä 6:sta 10:een mg/kg. Syynä muutokseen oli elintarvikkeiden seleenipitoisuuden ja väestön seleeninsaannin väheneminen. Tämä johtui siitä, että lannoitteiden seleenimäärää vähennettiin vuonna 1990 ja lannoitteiden käyttö väheni.

Lannoitteiden seleenipitoisuuden suureneminen on selvästi nostanut rehujen, peruselintarvikkeiden ja ihmisen seerumin seleenipitoisuuksia sekä väestön keskimääräistä seleeninsaantia. Vuoden 1998 jälkeen kotimaisen vehnän ja rukiin keskimääräinen seleenipitoisuus on kaksinkertaistunut. Vaihtelu on kuitenkin suurta, <0,010–0,260 mg/kg ka. Maidon seleenipitoisuus on noussut noin 30 % ja se on välillä 0,20–0,26 mg/kg ka. Myös lihan seleenipitoisuus on noussut 30–50 %. Seleenilannoituksen lisäys on vaikuttanut vain vähän kananmunien, sianlihan ja kasvisten seleenipitoisuuksiin.

Seleenilannoituksen vähentäminen vuonna 1990 laski suomalaisten seerumin seleenipitoisuuden 1,10 µmol/l vuonna 1999. Pitoisuus oli kuitenkin yhä suurempi kuin keskimääräinen pitoisuus Euroopassa, mutta yleisesti pienempi kuin Yhdysvalloissa ja Kanadassa. Vuoden 1998 jälkeen seerumin ja veren keskimääräinen seleenipitoisuus on jälleen alkanut nousta lannoitustason nostamisen seurauksena. Seerumin seleenipitoisuus on nyt noin 1,4 µmol/l.

Väestön keskimääräinen seleeninsaanti vuonna 2001 oli noin 20 % suurempi kuin vuonna 1999. Saanti on 0,08 mg/vrk energiatasolla 10 MJ. Suomessa seleeninsaanti on nyt riittävällä ja turvallisella tasolla ja yleisesti suurempi kuin monissa Euroopan maissa. Se vastaa hyvin RDA- ja DRI-suosituksia (0,055 mg/vrk). Tärkeimpiä seleeninlähteitä ovat maito ja maitotuotteet sekä liha ja lihatuotteet, joista saadaan lähes 70 % seleenistä.

Avainsanat: seleeni, saanti, lannoitteet, maaperä, elintarvikkeet, vilja, maito, liha, seerumi, ravinto, ravitsemus

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Contents

1 Introduction

For climatic and geochemical reasons Finland is one of the low selenium (Se) regions in the world. In the 1970s, the results of the extensive ‘Mineral Element Study’ (Koivistoinen 1980) showed that the Se content in domestic agricultural products was very low. Attention was also given to the possible health effects of the very low levels of Se in the average Finnish diet. To improve the quality of Finnish foods and to increase the Se intake of the population, an official decision was made in 1984 to supplement compound fertilizers with Se.

In 1983, the Ministry of Agriculture and Forestry appointed a selenium working group and gave it the of organizing an Se monitoring program. The Se follow-up study was initiated in 1984, and the effects of the Se content of fertilizers on soils, basic foods, feeds, human serum and Se intake have since been monitored regularly. During this period, the Se level in fertilizers has been changed twice. In 1984-1990 two supplementation levels were used: 6 mg Se/kg for compound fertilizers intended for fodder and hay production and 16 mg Se/kg for fertilizers intended for grain production. In 1990, the system was simplified by applying the 6 mg Se/kg level to all compound fertilizers, and in 1998, the Se supplementation level was raised to 10 mg Se/kg. The increment in the supplementation level was considered necessary because of the decreasing trend in the Se content of foods, feeds, and human serum and the decreasing Se intake. This was due to the lower Se supplementation level and decreased use of fertilizers. Environmental rules and restrictions in the use of phosphorus have decreased the use of compound fertilizers, and so the amount of Se per hectare has decreased continually.

The Se monitoring program has been effective in measuring the effects of Se fertilization and evaluating the Se intake. A large amount of data has been collected and published during the 19-year period of Se supplementation.

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2 Materials and methods

The participating organizations carried out sample collection and Se analyses according to their own protocols and methods. Soil analyses were performed by Viljavuuspalvelu Ltd., and fertilizer and feed analyses at the Plant Production Inspection Centre. Se monitoring of cereals and foods (excluding meat) was performed at MTT Agrifood Research Finland. Meat and liver were analyzed at the National Veterinary and Food Research Institute. Human studies were performed at the National Public Health Institute.

2.1 Samples

Cereals. Flours and silo samples of bread grain cereals (rye, spring and winter wheat) were obtained from the largest commercial mills in Finland: Oy Karl Fazer Ab, Raisio Group and Helsingin Mylly Oy. The flour samples were taken directly from the production lines at the mills. The farm samples of wheat, rye, barley, and oats were obtained from the Plant Production Inspection Centre Grain Laboratory, which has been collecting samples from farmers for its grain quality monitoring project since 1966.

Bread, milk, cheese, fish. Samples of bread, milk, cheese, Baltic herring and eggs were purchased 4 times a year from 8 retail food stores. The sampling areas varied: the samples were taken from Forssa every sampling period while the cities of Helsinki, Turku and Tampere alternated as a pair with Forssa (4 stores per city). The samples were pooled in pairs to make four analyzable samples per sampling period. This amounted 24 samples per food item annually. Rainbow trout, potato and white cabbage were collected only once each year. Occasionally some other food items such as carrot, strawberry, onion, and broccoli was also collected. Samples were freeze dried, homogenized with a blender, and stored in a freezer prior to analysis.

Meat and liver. Samples from pig and cattle were obtained from a Finnish slaughterhouse. The samples were collected regularly every month according to an annual residue control plan, from animals selected randomly from the slaughter line. The samples were packed separately and sent to the laboratory within 24 h in temperature-controlled chambers containing coolant canisters frozen before dispatch.

Fertilizers. The Se content of fertilizers was analyzed at the Plant Production Inspection Centre in the form of normal control inspections set by the fertilizer legislation. In 2000–2001, 33 samples were analysed.

Feeds. Samples were collected according to the European Commission regulation 138/1998/appendix 1.

Human blood and tissues. Serum samples have been obtained annually from healthy subjects in urban Helsinki (n=30-35) since 1984, and both serum and whole blood samples from rural Leppävirta (n=35-45) since 1985. The participants in Helsinki in 2000-2001 consisted of the members of staff of the National Public Health Institute, while those residing in Leppävirta were mostly the same subjects recruited in 1985 for the purposes of observing their Se status. Their ages in the year 2000 ranged from 26 to 62 years in Helsinki and 25 to 77 years in Leppävirta. All subjects filled in a questionnaire regarding their health status and use of dietary supplements (Se) on each blood-sampling occasion. Annual blood sample was taken after an overnight fast drawn into a vacuum tubes and stored at -20ºC before analysis (within 6 months). Sampling of other human tissues, toenails, and autopsy liver samples is described in Alfthan 1997 and Aro et al. 1998.

2.2 Analytical methods

Cereals, milk, cheese, vegetables, fish. Se was analyzed by an electrothermal atomic absorption spectrometry (ETAAS) method (Kumpulainen et al. 1983) accredited in 1999. The dried samples were digested in a mixture of concentrated HNO_3, $HClO_4$ and H_2SO_4. Se was reduced to Se(IV) with hydrochloric acid, chelated with ammonium pyrrolidine dithiocarbamate (APDC) and extracted into methyl isobuthyl ketone (MIBK) for AAS measurement. Certified (Table 1) or in-house reference materials of a suitable matrix were analyzed in every batch of samples. The Finnish Centre of Metrology and Accreditation has accredited the selenium method.

Meat and liver. Two methods were used for analyzing meat and liver: a fluorometric method (until 1990) and a hydride method (between 1990 and 1995) with digestion in a muffle furnace (Venäläinen et al. 1997). The samples were digested in a microwave oven (Questron Q-Wave 1000). The Se concentration was measured by hydride atomic spectrometry at 196.0 nm (Perkin-Elmer 4100 PC, electrodeless discharge lamp (EDL), EDL Power Supply (system 2), FIAS 200, AS90 autosampler. Until 1996, the same technique was used but with different instrumentation (Venäläinen et al. 1997). The Finnish Centre of Metrology and Accreditation has accredited the selenium method. The laboratory fulfils the requirements of the SFS-EN ISO/IEC 17025 standard.

Certified reference materials (Table 1) and blanks were analyzed together with each sample series. Calibration was continuously checked using standard samples with different concentrations. The recoveries in different sample materials were 80-105%. The amounts added were selected so as to be close to the amounts normally found in meat and liver.

Table 1. Results of the certified reference materials analyzed in the National Veterinary and Food Research Institute and MTT Agrifood Research during 2000-2001.

Certified reference material	Certified value mg/kg dw	Present results mg/kg dw	n
National Veterinary and Food Research Institute			
BCR 184 Bovine muscle	0.183 ± 0.012	0.176 ± 0.015	35
BCR 185R Bovine liver	1.68 ± 0.14	1.67 ± 0.021	5
NIST Bovine liver	0.73 ± 0.06	0.73 ± 0.03	31
MTT Agrifood Research Finland			
ARC/CL Wheat flour	0.057 ± 0.005	0.055 ± 0.003	24*
ARC/CL Milk Powder	0.082 ± 0.008	0.075 ± 0.006	12*
Dorm-2 Dogfish muscle	1.40 ± 0.09	1.41 ± 0.01	5*
NIST Bovine liver	0.73 ± 0.03	0.75 ± 0.02	9*
ARC/CL Diet	0.181 ± 0.017	0.183 ± 0.03	3*

* number of sample batches; may include more than one analytical batch (more than one reference)

Fertilizers and feeds. Se was analyzed by the hydride generation atomic absorption spectrometry (HGAAS) method. Feed samples were wet digested and fertilizers diluted to acid solution prior to the AAS measurement. The method used was MKO number P08261.

Soils. At Viljavuuspalvelu Ltd. soil Se was analyzed by an in-house method. Se was extracted with boiling water (1:3 30 min) and dry ashed with the aid of $Mg(NO_3)_2$. Concentrations of Se were measured by atomic absorption spectrometry using FIAS technique.

Serum, whole blood and tissues. Serum Se was determined by an ETAAS method (Alfthan & Kumpulainen 1982). The precision between series was 4.4% and the accuracy was, on average, within 5% of analyses of external quality-assurance serum standards (Alfthan et al. 2000). Whole blood, toenails and liver Se were determined by an acid-digestion fluorometric method (Alfthan 1984). The precision of the method between series was 3.5%, and the mean bias compared with external quality assurance samples was 1%.

2.3 Quality control

The laboratories participating in Se monitoring program used their own quality control schemes in the sampling and the Se analyses. Most of the laboratories participate regularly in international comparison tests. In addition, the

University of Helsinki has organized interlaboratory comparison tests 21 times during the years 1985-1997, and this has included laboratories monitoring Se. MTT Agrifood Research Finland organized these tests in 1999 and 2001. The samples (4-5) represented different food and feed matrixes, with Se concentrations ranging from very low to high.

In 1999 and 2001 the z-scores of the laboratories varied in the range 0-1.8. The statistical analyses proved that there were no significant differences in the Se levels between the Se monitoring laboratories, and the results from different laboratories are comparable with each other.

3 Selenium in agricultural soils

In 1970s and 1980s, few basic studies on the Se content of Finnish agricultural soils were performed (Koljonen 1975, Sippola 1979, Yläranta 1985, Sillanpää & Jansson 1992). Yläranta (1985) found a mean total Se content in the plough layer of 0.21 mg/kg. The coarse mineral soils studied by Yläranta (1985) contained lower Se concentrations than clay soils, and the highest concentrations (1 mg/kg) were found in organogenic soils. The total Se content in Finnish soils is not exceptionally low. However, for climatic and geochemical reasons Se is poorly available to plants. The amount of hot-water extractable Se can be used as an indicator Se availability to plants. About 4% (range 1.5-10.2%) of the total Se has been found to be extractable in hot water from various soil types (Yläranta 1985).

Viljavuuspalvelu Ltd. has been analyzing hot-water extractable Se from farming samples. Since Se supplemented fertilization began, the number of samples received has decreased considerably, and so the selenium working group had a soil Se study carried out by Viljavuuspalvelu Ltd. in 1990 (Ministry of agriculture and Forestry 1994). The mean hot- water extractable Se content of the agricultural soils (n=450) was in that study 0.006 mg/l. According to Sippola (1979), the mean total Se content of soil samples was 0.201 ± 0.019 mg/kg (n=250, range 0.005-1.241 mg/kg). Clay and mull soils contained higher Se concentrations than coarse mineral soils. MTT Agrifood Research Finland has monitored soil quality in 1998 (Mäkelä-Kurtto & Sippola 2002), and found that mean soluble (extracted to hot water) Se content of soils in 1998 was 0.010 ± 0.005 mg/l (n=705), with concentrations decreasing northwards. The mean result obtained in 1990 is within the confidence limits of the results by Sippola (1979) and Mäkelä-Kurtto and Sippola (2002). Therefore, on the basis of these two large materials, it seems that there is no marked increase in the concentration of hot-water extractable Se in agricultural soils of Finland.

Recently the number of Se analyses by Viljavuuspalvelu Ltd. has been as low as 9-12 samples per year. In 2000 the hot-water extractable Se content varied between <0.01 and 0.12 mg/l (n=10), and in 2001 between <0.01 and 0.13 mg/l (n=9). The present results and the literature data indicate that Se fertilization has not increased the amount of hot-water extractable Se in agricultural soils. However, the total Se content of soils has not been monitored during the Se fertilization program. Utilization of fertilizer Se by the crop is usually <10%. It can be assumed that most added Se not taken up by the crop is immobilized in the soil. A rough calculation suggests that since 1984 the content of total Se has increased about 70 g ha^{-1}. If all of it remains in the soil and it is distributed evenly throughout a 20 cm thick plough layer, total Se in Finnish agricultural soils has increased on average about 20 %.

4 Selenium in fertilizers

4.1 History

The addition of selenium (Se) to all the main granular compound fertilizers was started in Finland in 1984 by the decision of the Ministry of Agriculture and Forestry (21. 3. 1984). Under the decision, it was obligatory to add 16 mg/kg of Se in the form of sodium selenate to fertilizers for cereals and horticultural crops, and 6 mg/kg to fertilizers for grassland. In 1991, the amount of Se was lowered to 6 mg/kg in all compound fertilizers. There were many reasons for this. For instance, some Finnish environmental activists had claimed that the increase in algae in lakes and river was a result of the addition of Se to fertilizers. In official control inspections, some crop samples were found to contain higher amounts of Se than was intended. The authorities did not wish to take any risks and insufficient studies have so far been carried out. After six years it was noticed that the selenium intake of people and especially animals had decreased so much that some veterinarians even reported diseases caused by Se deficiency in different parts of the country. Another reason for this was that the usage of fertilizers decreased considerably. Therefore, the addition of Se to inorganic compound fertilizers was increased to 10 mg/kg in 1998.

Today, we have more experience and knowledge of Se fertilizing. It seems that fertilizer Se given in the form of sodium selenate is effective on plants, but only in the first summer, as it quickly turns into an unavailable and insoluble form in the soil. Se fertilizing needs to be carried out every year; bigger concentrations in fertilizers do not help. For the same reason, no leaching of Se from the soil is detected. Annual dosage to soil is normally under 5g/ha, and only part of it leaves to the soil. Long-term environmental risks are not likely. In normal farming it is possible to make the use of Se more efficient by using new methods. ‘Precision’ fertilizing helps to achieve more exact dosages of nutrients, including Se, to plants, minimizing leaching and other environmental risks. For instance, ‘placement’ fertilizing has been used in Finland for over 30 years. The fertilizer is applied to the soil near the seed row with a special combined sowing/fertilizing machine in such a manner that the plant roots can use the nutrients and the Se better than in broadcast spreading. The method gives bigger yields with lower fertilizing needs, so that farmers can save money and reduce adverse environmental effects. It also helps to maintain an optimum level of Se in plants with the lowest possible addition of Se to fertilizers.

4.2 Monitoring

The permitted tolerance given in the Ministry of Agriculture and Forestry decision on Se in fertilizers is ± 50% (46/1994). It is the relative deviation of measured Se content from the nominal content, which is at present 10 mg/kg. The tolerance is intended to accommodate variations in manufacturing, sampling and analyses. The permitted absolute range of Se in compound fertilizers is 5-15 mg/kg.

The average result obtained in 2000-2001 Se monitoring was 9.9 mg/kg the range 5.0-13.0 mg/kg and the standard deviation 1.8 mg/kg. All 33 samples analyzed in our inspections during 2000-2001 were within the permitted tolerances.

On the basis of the analyses made in 2000-2001 we can state that the addition Se to fertilizers has been precise and trouble-free. It is a safe and inexpensive way to ensure an adequate Se intake for domestic animals and humans in Finland, where the soil does not naturally contain a sufficient amount of this important trace element.

5 Selenium in feeds

5.1 Selenium in commercial feeds and silage

Besides organic Se, which comes from cereals and other fodder ingredients, animal feeds can be supplemented with inorganic Se (sodium selenite, sodium selenate). The companies that produce Se supplemented feeds must take certain measures required by law. The maximum Se content of feeds is 0.5 mg/kg calculated as full-feed. The maximum Se content of mineral feed is 20 mg/kg (Ministry of Agriculture and Forestry regulation 125/1998). Mineral feeds have to contain such large quantities of mineral ingredients that they cannot be used in the same way as full-feeds, but must be mixed with other feeds according to the instructions. The Plant Production Inspection Centre has monitored the Se levels in commercial feeds (Table 2) andsilage (Table 3). The number of feed samples in 2000-2001 was 111. 40 feed

Table 2. Selenium (Se) content of feeds in 2000 – 2001 by samples. Values exceeding the maximum levels are in **bold** type.

Product	Year	n	Selenium content mg/kg (if underlined mg/l; double line mg/ml)											
Poultry feeds	2000	4	**0.55**	0.33	0.35	1.9								
	2001	5	0.47	0.47	**0.5**	0.4	**0.58**							
Pig feeds	2000	10	**0.62**	0.33	0.39	2.2	2.2	2	1.7	**4.3**	**2.8**	1.4		
	2001	6	**0.6**	0.49	1.8	1	**0.63**	4.5						
Cattle feeds	2000	7	0.36	0.68	0.27	0.16	0.76	**0.5**	1.6					
	2001	11	**0.54**	**0.6**	**0.53**	**0.3**	0.63	0.5	0.5	0.5	0.6	0.65	0.8	
Mixed mineral feeds	2000	24	11	**6.5**	**9**	17	18	18	17.5	**23**	19	**24**	17	6
			8.3	16	17	13	**22**	20	20	5.4	17	18	2.3	5.6
	2001	16	**7.8**	20	**23**	**21**	20	15.2	1.8	**22**	**5.5**	7.0	10	20
			19	330	219	200								
Feed additives	2000	11	11.2	**0.011**	250	19	20	0.020	0.020	0.009	<0.1	11.0	4.3	
	2001	-												
Fish feeds	2000	10	**0.8**	**1.5**	**1.6**	**1.2**	**2.3**	**1.3**	**2.4**	**0.8**	**0.93**	**0.73**		
	2001	5	**0.8**	**1.2**	**1.2**	**1.6**	0.7							
Dog and cat feeds	2000	-												
	2001	2	<0.05	**0.56**										
Total	2000	66	Remarks 22 (33 %)											
	2001	45	Remarks 18 (40 %)											

n = number of samples

Table 3. Se content of silage (analyzed in the Plant Production Inspection Centre). Samples collected in 2000 and 2001.

Year 2000 Silage	n	Se content mg/kg dw mean	 range
I harvest, spring	13	0.51	0.22 – 0.81
II harvest, summer	8	0.28	0.15 – 0.46
III harvest, fall	12	0.33	0.22 – 0.47
Organic cultivation	8*)	0.04	0.01 – 0.08

*) I harvest n=1. II harvest n=3. III harvest n=4

Year 2001 Silage	Se content mg/kg dw	Fertilization information
I harvest, spring	0.45	Y-2 for grasses 450 kg/ha
n=9	0.40	NK for grasses 500 kg/ha
	0.28	Sludge 30 tons, NK for grasses 200 kg/ha
	0.37	-
	0.83	-
	0.75	-
	0.27	PK for grasses 420 kg/ha
	0.20	Sludge 25 tons, NK 265 kg/ha
	0.26	-
II harvest, fall n=3	0.05	Sludge 15 tons, Y-5 140 kg/ha, cutting, 192 kg/ha N-fertilizer, this cutting
	0.21	-
	0.23	Y-6 300 kg/ha, cutting, NK 250 kg/ha this cutting
Organic cultivation (n=3)		
I harvest, spring	0.05	Urine 20 tons nitrogen 20 kg/ha
I harvest, spring	0.03	20% clover
II harvest, fall	0.11	-
Mean	**0.30**	
Standard deviation n=15	**0.24**	

samples had Se contents exceeding the permitted value, and 14 of these were in fish feed. The high Se contents came from the fish meal used in the production of fish feeds; feeds did not contain supplemented Se. This reflects the difficulties of the feed industry in estimating the amount of organic Se that comes from feed ingredients. However, even if the Se content is occasionally higher than the permitted levels, there is little danger of overdose as the bio-availability of inorganic Se is poor.

The mean Se content of silage in 2001 (from fertilization with inorganic fertilizers) was 0.30 ± 0.24 mg/kg dw. In organic cultivation the Se content of silage was low, about 10 times lower than in conventional cultivation. Se contents in silage made in Viljavuuspalvelu Ltd. are presented in Table 4.

Table 4. Se content in silage in 2000 and 2001 (analyzed in the Viljavuuspalvelu Ltd).

Silage	**Se content mg/kg dw**		
	n	**mean**	**range**
Predried I harvest	36	0.24	<0,02-0.44
Fresh preserved I harvest	15	0.22	<0.02-0.57
Predried II harvest	6	0.19	<0.02-0.38
Fresh preserved II harvest	10	0.19	<0.02-0.27
Predried III harvest	8	0.12	0.03-0.28
Fresh preserved III harvest	3	0.15	0.02-0.23

5.2 Selenium in fodder oats and barley

Se content of fodder oats and barley has been monitored since 1984. The mean Se content was at its highest in 1988-1990: 0.23-0.26 mg/kg dw (Ministry of Agriculture and Forestry 1994). However, the variation in Se content has been large (0.002-0.84), as the samples are single unpooled farm samples and the growing conditions vary considerably. Since 1990, the Se content in oats and barley has decreased gradually. In 1998 the mean Se content of oats was 0.05 ± 0.03 mg/kg dw and of barley 0.04 ± 0.03 mg/kg dw (Eurola et al. 2000). The increment in the Se supplementation level has clearly affected the Se content of oats and barley (Fig. 1). In 1999-2001 the mean Se content of oats and barley doubled, fluctuating between 0.107 and 0.132 mg/kg dw, range <0.010-0.330 mg/kg dw. It seems that Se concentrations are stabilizing to a mean level of 0.100-0.140 mg/kg dw, which is close to the original target level of 0.1 mg/kg dw in cereals (Eurola & Hietaniemi 2000).

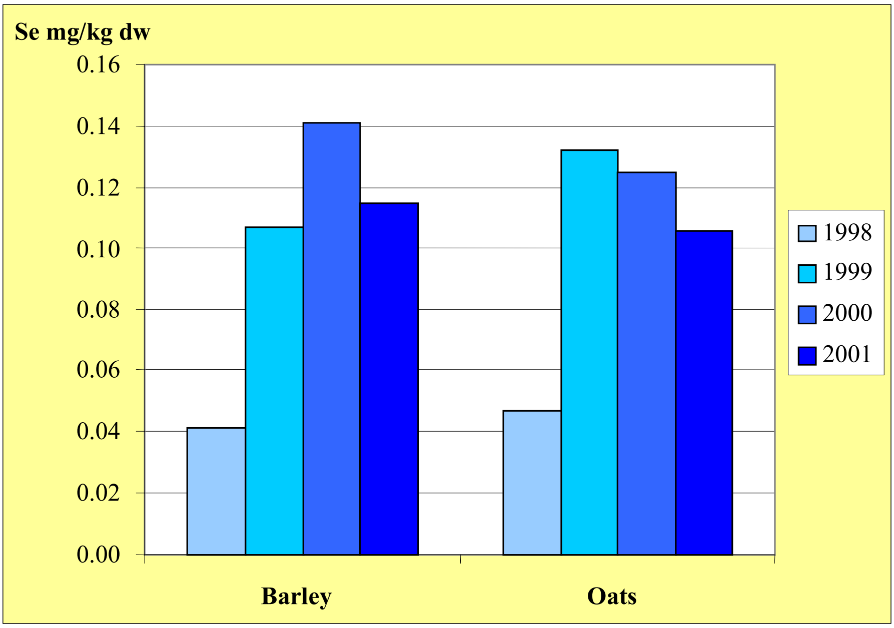


Fig.1. Mean Se content of fodder oats and barley in Finland during 1998-2001.

6 Selenium in foods

6.1 Introduction

Since 1984 the Se content of basic foods has been monitored regularly, first by the University of Helsinki and, since 1998 by MTT Agrifood Research Finland (cereals, milk vegetables, fish) and the National Veterinary and Food Research Institute (meat). The Se supplementation level has been revised twice since 1984. In 1990, the system simplified and converted to the uniform Se fertilization rate: 6 mg Se/kg. This change clearly decreased the Se content of most basic foods (Ekholm et al. 1994). However, in 1998 the Se supplementation dose was increased to 10 mg/kg, as the Se intake had declined due to the decreased use of fertilizers and lower level of Se supplementation since 1990. The environmental regulations restrict the use of fertilizers.

Se content of organically grown foods have been studied during 2001-2002. Results have been reported by Ministry of Agriculture and Forestry (2003).

6.2 Cereal grains, flour and bread

In 1984 when Se fertilization was started, the primary objective was to raise the Se content of domestic cereal grains to an approximate level of 0.1 mg/kg dw (Eurola & Hietaniemi 2000). The effect of Se fertilization was distinct. Already in the 1985 growing season the Se content of spring wheat increased to 0.23 ± 0.10 mg/kg dw and it remained at the mean level of 0.2-0.3 mg/kg dw during the years when the supplementation level for cereals was 16 mg Se/kg fertilizer (Ekholm 1997). Se fertilization did not affect winter cereals (winter wheat and rye) as much as spring cereals; the Se levels remained mainly around 0.04-0.05 mg/kg dw (Table 5, Fig. 2). The reason for this is the different fertilization and cultivation practices. In the fall, only a slight dose of Se supplemented fertilizer for winter cereals was applied and in spring plain nitrogen fertilizer was used. Moreover, selenate is easily reduced in wet winter conditions and bound to the soil constituents (Eurola et al. 1990, Ekholm 1997). During the years of lower supplementation level in 1990-1995 the Se content of spring wheat decreased to 0.08 mg/kg dw, whereas in winter cereals Se levels were not significantly affected (Ekholm 1997). In 1996, Se was also added to nitrogen fertilizer. This has raised the Se content of winter cereals to about 0.1 mg/kg dw (Fig. 2).

Table 5. Se contents of wheat and rye grains in Finland during 1998-2001.

Year	Selenium content mg/kg dw					
	n	Spring wheat	n	Winter wheat	n	Rye
Silo samples from mills						
1998	3	0.076 ± 0.011	3	0.052 ± 0.010	2	0.066 ± 0.000
1999	4	0.130 ± 0.010	2	0.097 ± 0.025	1	0.120
2000	3	0.160 ± 0.014	2	0.130 ± 0.008	2	0.110 ± 0.006
2001	4	0.160 ± 0.050	3	0.091 ± 0.022	3	0.130 ± 0.027
Farm samples						
1999	13	0.150 ± 0.021	13	0.120 ± 0.015	22	0.130 ± 0.083
2000	14	0.190 ± 0.082	14	0.110 ± 0.042	12	0.110 ± 0.048
2001	21	0.130 ± 0.080	14	0.140 ± 0.051	15	0.084 ± 0.063

The periods of different Se fertilization levels are clearly seen in the Se content of cereal grains. The Se content of wheat and rye has doubled since the last increment in the Se supplementation level in 1998 (Fig. 2, Table 5). However, the variation in the farm samples was large: in 2001 spring wheat 82-230, winter wheat <10-260 and rye <10-190 mg/kg dw. The major sources of such variation are the amount of fertilization, the soil type, other conditions and the cultivar. The silo samples from commercial mills represent large amounts of grain (0.04-0.6 million kg) and give a good overview of the Se situation in wheat and rye (Table 5).

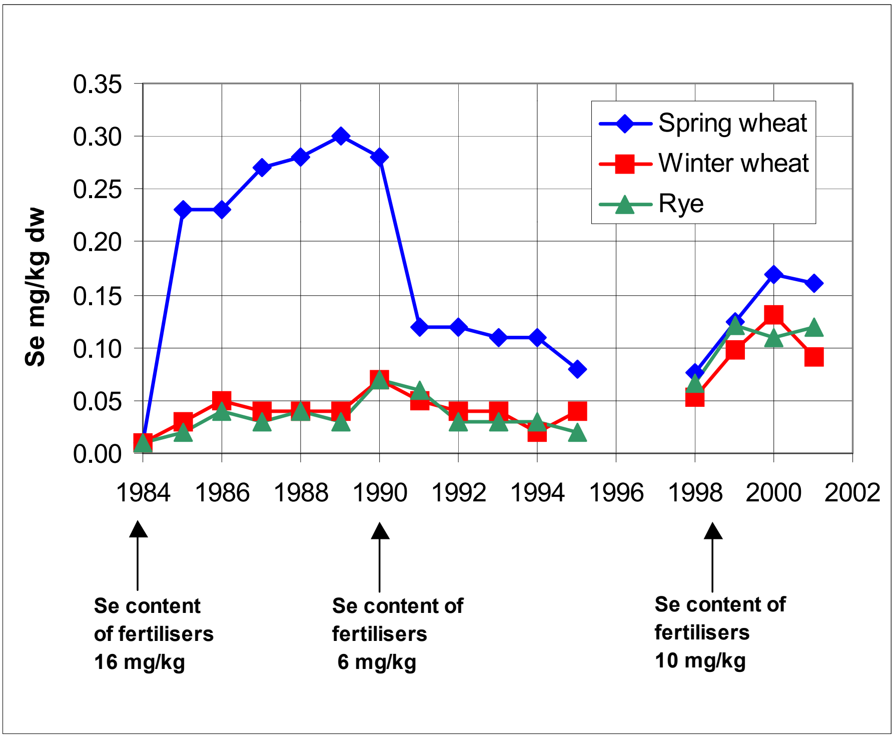


Fig. 2. Trends in the Se content of wheat and rye grains during 1984-2001 (silo samples from commercial mills).

The Se contents of the raw materials determine the Se levels in flours and breads. Se fertilization has increased the Se content of wheat and rye flours 10-20-fold (Figs 3 and 4). However, the Se content of flours and breads does not necessarily correlate with the Se content of domestic grain, as it is also affected by the proportion of imported grain in milling. During years of serious crop failure when high-quality domestic grain is not available, the amount of imported grain in milling can be as high as 100%. In the 1980s, most of the imported wheat was of North American origin, and generally had a higher Se content than Finnish wheat (Eurola et al. 1990). The origin of imported rye has been more variable. Recently, most of the imported grain has been of European origin, where the Se content is generally lower than in Finnish grains. In 1998-1999, the Se content of flours and breads was significantly lower than that of domestic grains due to grain imports from the EU region. The Se content increased immediately in fall 2000, when the new crop of domestic grain became available. In 2001, the mean Se contents of rye flour and bread were 0.073 ± 0.035 and 0.088 ± 0.026 mg/kg dw, and wheat flour and bread 0.122 ± 0.015 and 0.120 ± 0.013 mg/kg dw.

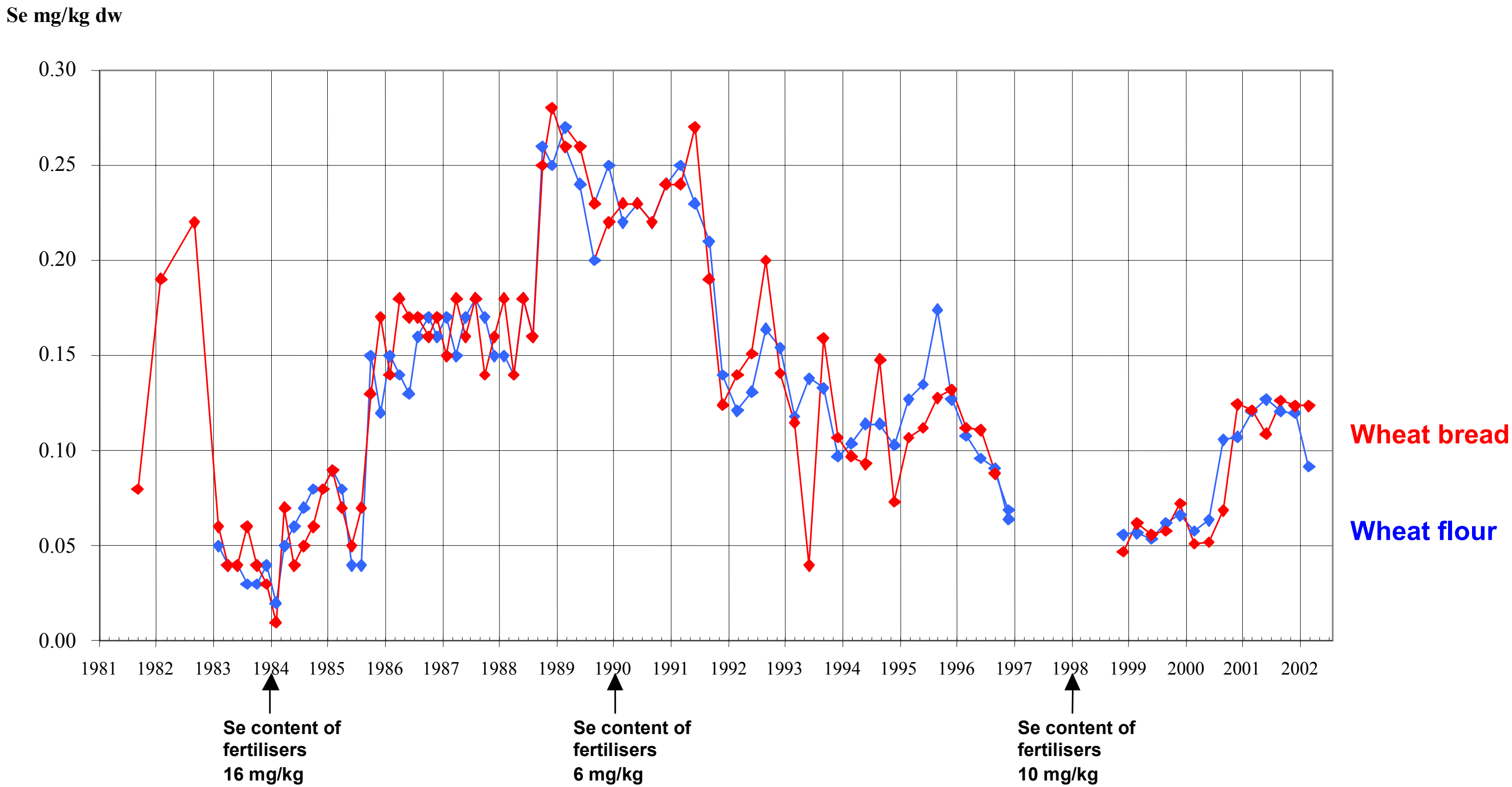


Fig. 3. Se content of wheat flour and bread in Finland during 1981-2001.

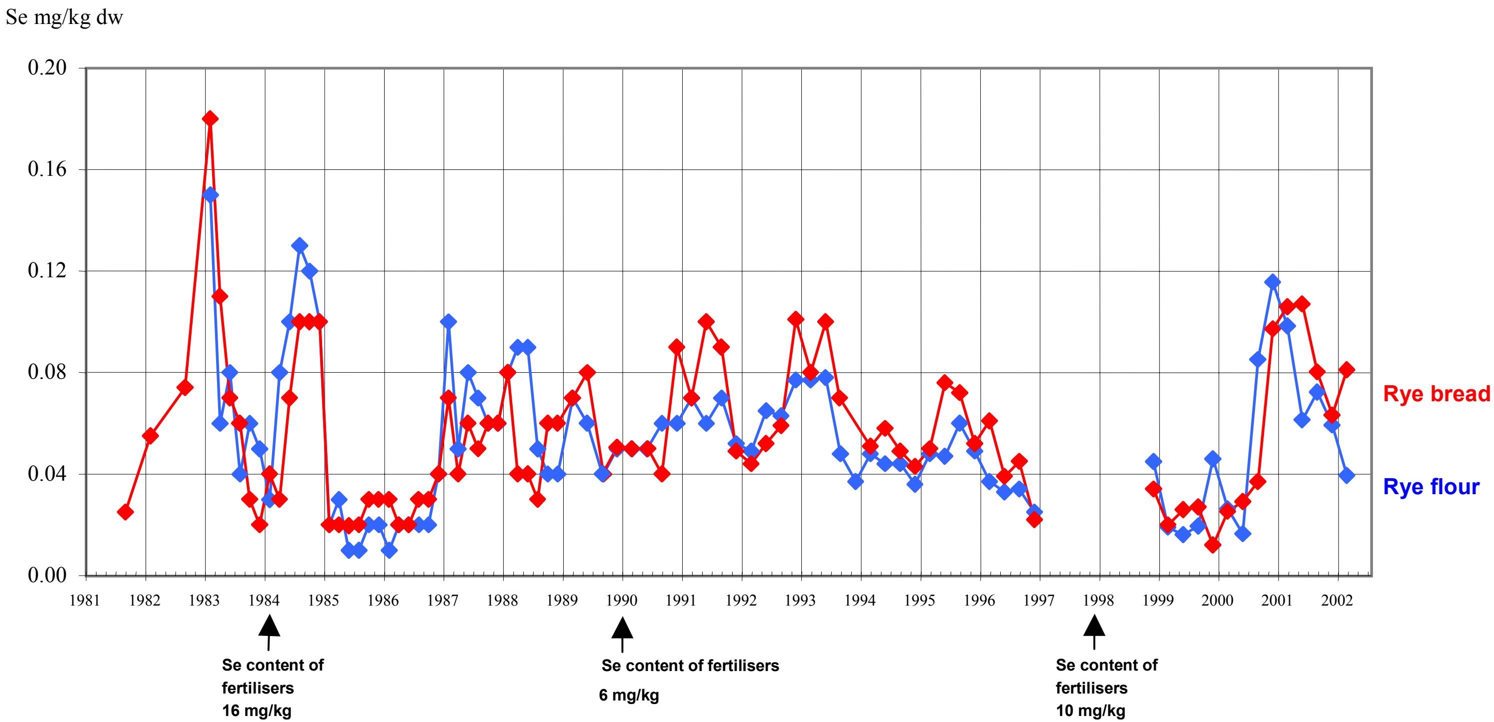


Fig. 4. Se content of rye flour and bread in Finland during 1981-2002.

6.3 Milk, cheese and eggs

The mean Se contents of standard milk, low-fat milk and Edam-type cheese were 0.235 ± 0.025, 0.264 ± 0.028 and 0.364 ± 0.044 mg/kg dw in 2001. When Se supplementation level was raised from 6 to 10 mg/kg fertilizer in 1998, the Se content of milk increased about 30% (Fig. 5). The ascending trend now seems to be stabilizing and the Se content of standard milk fluctuates between 0.20 and 0.26 mg/kg dw. The Se content of milk varies according to the season, being highest in the indoor feeding season and beginning to decrease in the outdoor feeding season (Fig. 5). Se contents are at their minimum in September.

Se in mainly bound to the protein constituents in milk. In cheese, the Se content is about 1.5 times higher than in milk, due to the higher protein content. In 1999, the increment in the Se fertilization level raised the Se content of cheese to 0.47 mg/kg dw. This was the maximum value obtained during the Se fertilization period and higher than the Se content of milk would presuppose. The reasons behind these unexpected Se contents remain indistinct. The summer of 1999 was exceptionally dry and warm, which may have affected the growing conditions of feeds and pastures increasing the availability of Se. In 2000-2001 the Se content of cheese settled at a level of 0.33-0.38 mg/kg dw (Fig. 6).

The trend in the Se content of eggs is shown in Fig. 7. The mean Se content of eggs were 1.10 ± 0.16 mg/kg dw in 2000 and 1.05 ± 0.13 mg/kg dw in 2001. The Se content are now slightly higher than during the lower Se fertilization period in 1990-1997.

6.4 Fish

Se fertilization has not affected the Se content of fish (Ekholm et al. 1990, Eurola et al. 1991). The mean Se contents of Baltic herring (*Clupea far. membra*) in 2000 and 2001 were 0.89 ± 0.15 and 0.77 ± 0.15 µg/kg dw, respectively (range 0.59-1.54 µg/kg dw). Se contents of cultivated rainbow trout (*Salmo gairdnerii*) were 0.52 and 0.41 µg/kg dw in 2000 and 2001 (range 0.34-0.59 µg/kg dw). Se content of fish correlates negatively with the fat content of the fish. Low-fat fish species generally contain more Se than more fatty species (Eurola et al. 1991). Moreover, the Se content of herring fluctuates according to the season, being usually at its highest in the summer when the fat content is lowest (Ministry of Agriculture and Forestry 1994, Eurola et al. 2000).

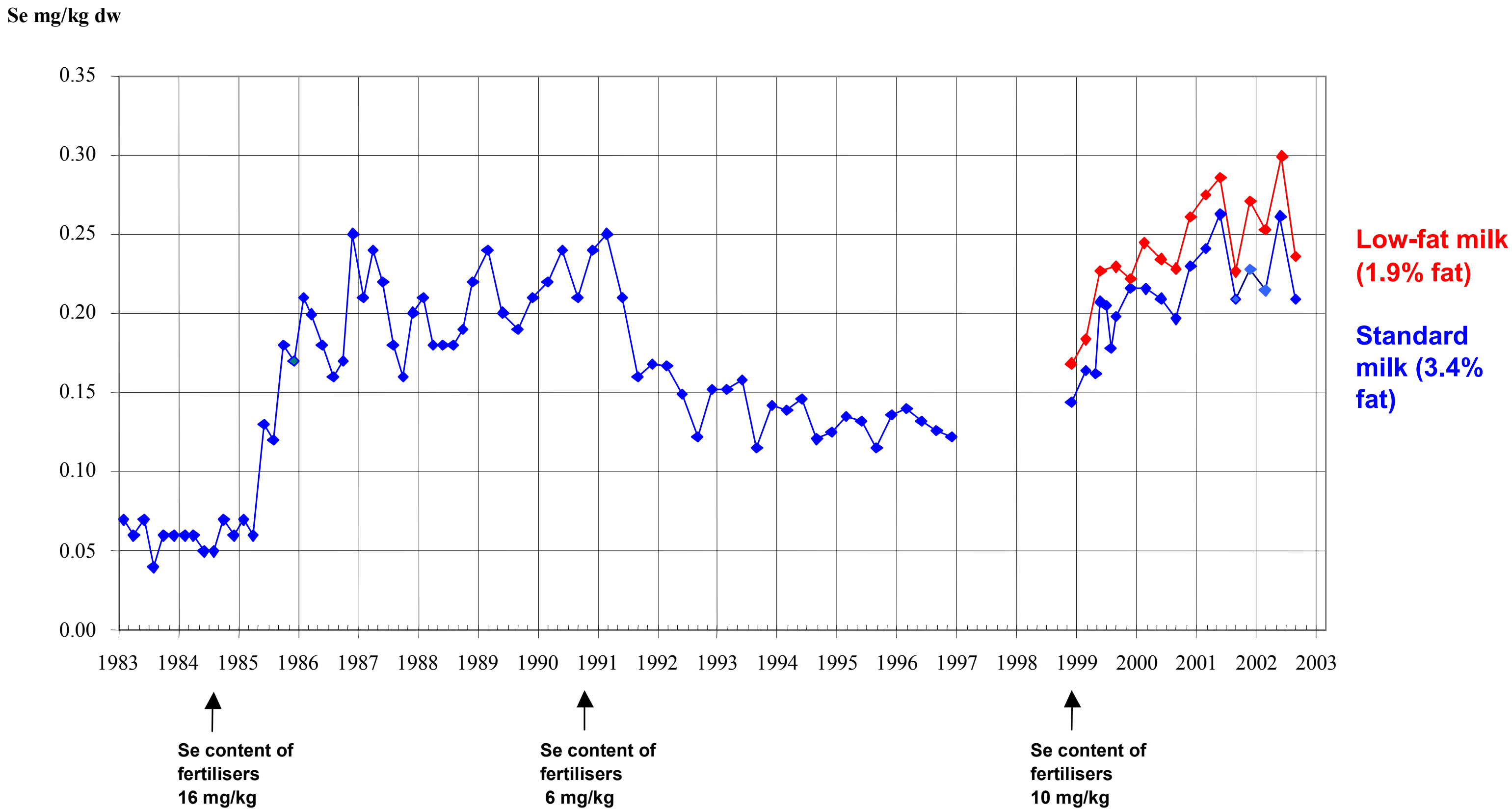


Fig. 5. Se content of milk in Finland during 1984-2001.

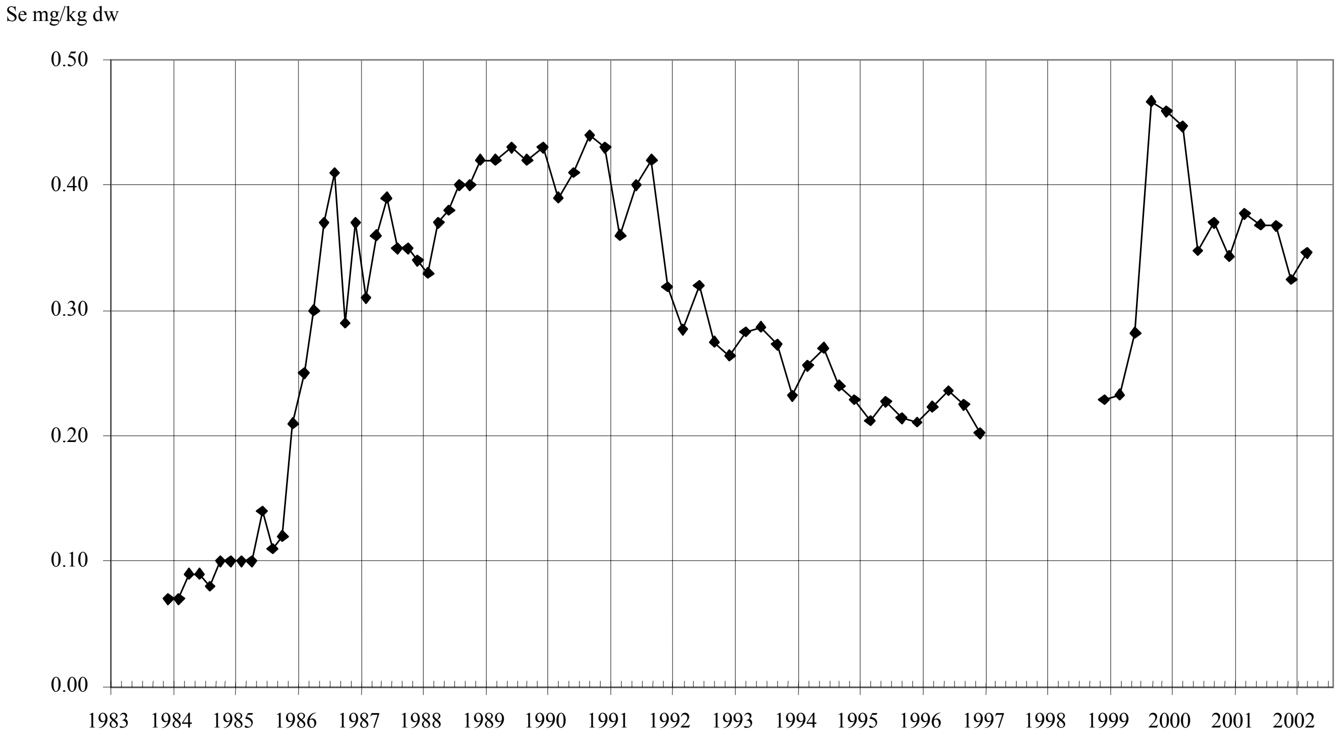


Fig. 6. Se content of cheese in Finland during 1984-2001.

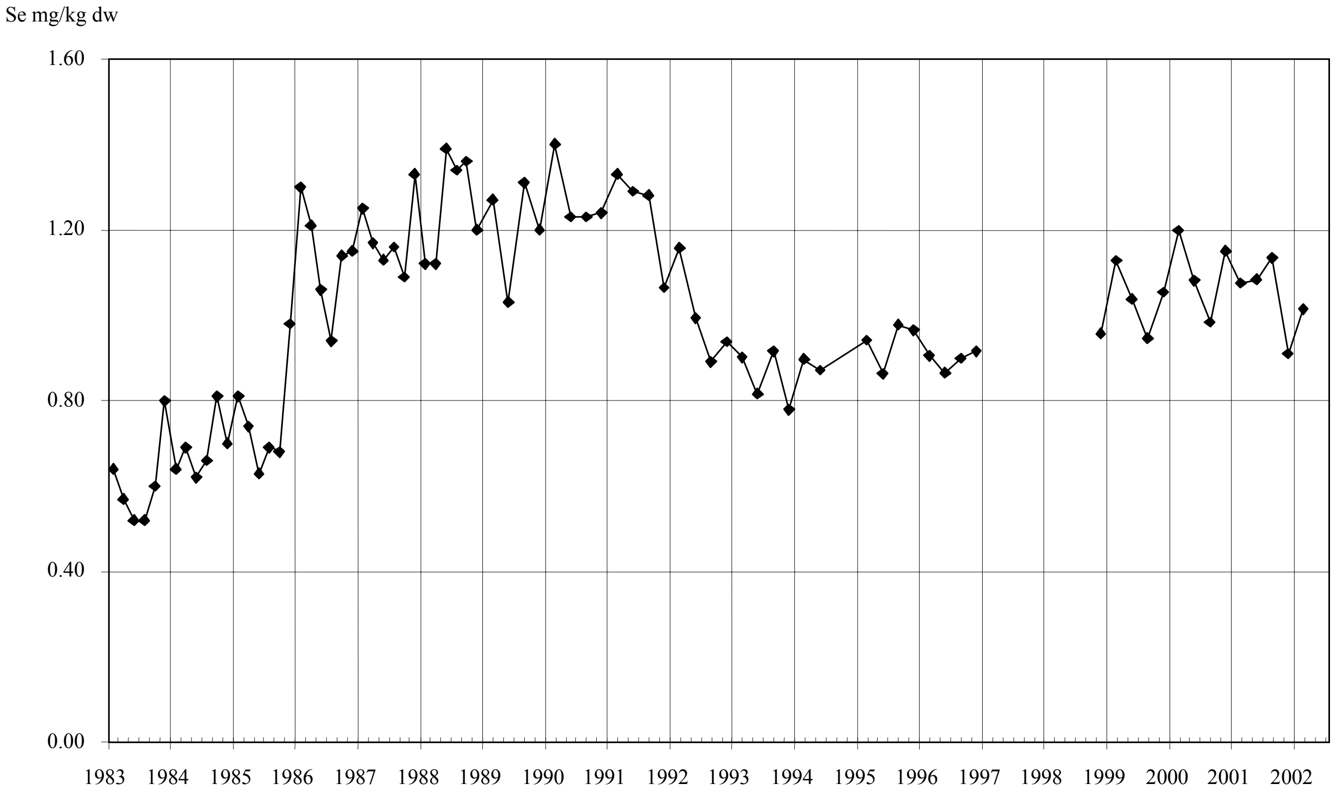


Fig. 7. Se content of eggs in Finland during 1983-2001.

6.5 Vegetables

In 2000-2001, the Se contents of potato, carrot and white cabbage were generally 2-4 times lower than in the late of 1980s (Table 6). Since 1990, the Se content of potato has decreased constantly throughout the period of the lower Se supplementation dose (Ministry of Agriculture and Forestry 1994). The decreasing trend now seems to have stopped, and slight increases in Se concentrations in vegetables have been detected. However, the amounts have been rather low and it is difficult to draw any reliable conclusions about the effects of the last increase in Se fertilization levels on vegetable foods.

Table 6. Selenium content of some vegetables in Finland.

Food	Year	n	Se content mg/kg dw	
			Mean	Range
Potato	1989*	12	0.11	0.05-0.20
	2000	4	0.026	0.016-0.042
	2001	4	0.031	0.020-0.055
Carrot	1989*	2	0.04	0.03-0.05
	2001	4	0.020	0.012-0.032
White cabbage	1989*	20	0.58	0.09-1.60
	2000	4	0.34	0.071-0.86
	2001	4	0.16	0.087-0.35
Strawberry	1989*	2	0.01	0.01-0.01
	2001	4	0.011	<0.010-0.022

* Eurola et al. 1991

6.6 Meat

The Se content of muscle and liver in Finnish pigs and cattle are shown in table 7. The Se fertilization began to affect Se contents in cattle in 1985, while the Se content in pig samples began to increase after 1985. (Figs 8 and 9). The reason for this difference between cattle and pigs was probably that the cattle were fed partly in outside pasture, whereas pigs were fed indoors only using feed that predated Se fertilization. The highest Se contents were reached in 1989, decreasing thereafter. The reduction in the amount of Se in fertilizers affected the Se content in muscle more slowly than in liver.

The increase in the amount of Se in fertilizer introduced in 1998 was reflected in the Se content of pig and cattle the same year. From the results of 2000 and 2001, it appears there is a rising trend in the Se content of pigs and cattle. The Se content of cattle in organic farming has been lower than in conventional production (Table 7). The insufficient Se intake from organic farming could cause deficiency diseases.

Table 7. Se contents of pig and cattle muscle and liver in conventional and organic production. Se concentrations on a dry weight basis are about 3 times the concentrations in fresh weight.

Year	Pig, muscle		Pig, liver		Cattle, muscle		Cattle, liver	
	n	Se mg/kg fw	n	Se mg/kg fw	n	Se mg/kg fw	n	Se mg/kg fw
Conventional production								
1974	40	0.11	60	0.45			60	0.10
1981	48	0.08	60	0.56	60	0.04	60	0.16
1985	23	0.12	23	0.49	26	0.07	26	0.28
1986	24	0.23	24	0.56	25	0.10	25	0.34
1987	30	0.26	30	0.64	40	0.16	40	0.42
1988	30	0.30	30	0.70	29	0.16	29	0.45
1989	34	0.29	34	0.73	35	0.21	35	0.51
1990	50	0.26	50	0.68	47	0.19	47	0.46
1991	30	0.26	50	0.62	30	0.16	30	0.40
1992	30	0.19	30	0.57	30	0.16	30	0.37
1993	30	0.19	30	0.56	30	0.14	30	0.35
1994	30	0.13	30	0.50	30	0.09	30	0.32
1995	31	0.15	30	0.52	30	0.10	30	0.28
1996	30	0.13	30	0.48	30	0.09	30	0.26
1997	32	0.13	29	0.46	31	0.09	29	0.27
1998	31	0.15	32	0.54	31	0.10	30	0.31
1999	30	0.14	33	0.51	29	0.08	30	0.33
2000	30	0.19	31	0.56	31	0.13	31	0.40
2001	38	0.20	44	0.56	30	0.16	31	0.46
Organic production								
1998	10	0.14	10	0.49	5	0.05	5	0.08
1999	3	0.08	3	0.36				
2001					10	0.06	10	0.23

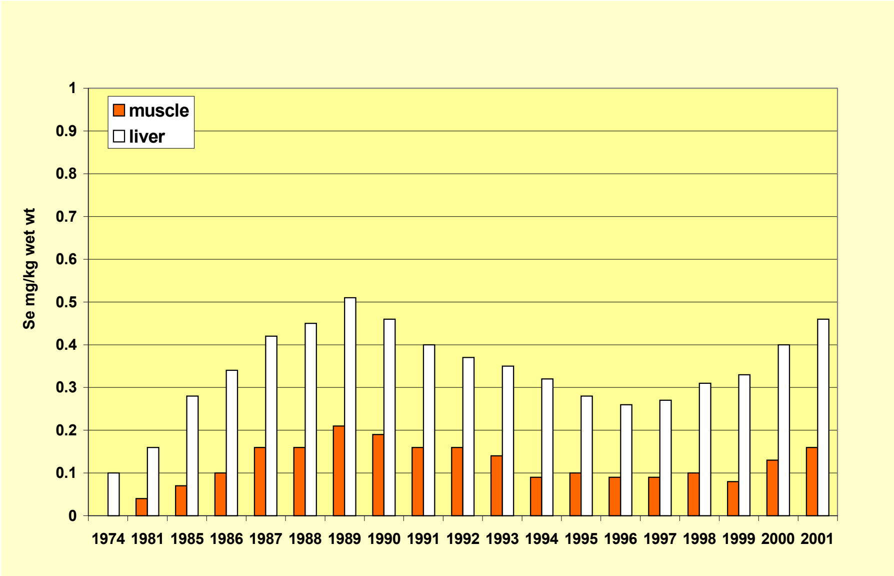


Fig. 8. Mean Se content of bovine meat and liver in Finland during 1974-2001.

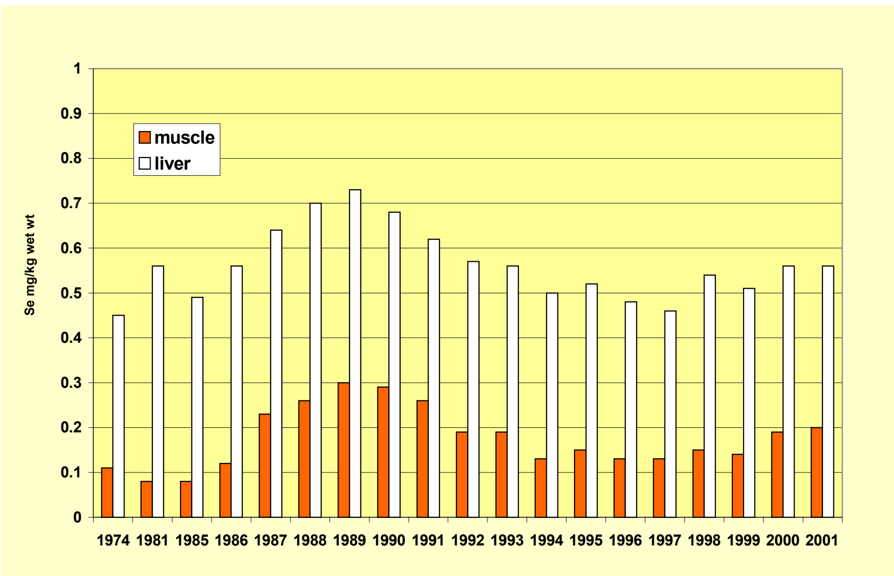


Fig. 9. Mean Se content of pig meat and liver in Finland during 1974-2001.

7 Selenium intake

The aim of Se fertilization was to guarantee adequate Se contents in foodstuffs, and to increase the human Se intake nationwide (Ministry of Agriculture and Forestry 1983). The Se intake was found to be extremely low in Finland during the 1970s (Varo & Koivistoinen 1980, Mutanen 1984). In the 1970s, during the years when all cereals used were of domestic origin the Se intake measured was one of the lowest ever reported internationally (Koivistoinen & Varo 1987).

The effects of Se fertilization on average daily Se intake have been monitored regularly since the beginning of the supplementation of fertilizers with Se in 1984. The estimation of average daily Se intake is based on Finnish food consumption statistics (preliminary 2001) at an energy level of 10 MJ and the Se contents of foods (Ministry of Agriculture and Forestry 2002). This method has been found to give reliable estimates compared with other methods such as urinary excretion. The method used overestimates the Se intake slightly. The statistics include the presence of waste (bones etc.). The difference between the method used and other estimation methods is not significant (Ekholm 1997).

The effects of Se fertilization on the average daily Se intake have been clearly seen from the very beginning. During the first Se fertilization period when two supplementation levels were in use, the average daily Se intake attained a level of 0.11-0.12 mg/d/10 MJ and remained nearly constant until 1990 (Fig. 10). The reduction in the amount of Se in fertilizers from 1991 to1997 was also clearly seen. The Se intake decreased from 0.12 mg/d to 0.09 mg/d between the years 1991 and 1992, and a minimum level was reached in 1999. (an average daily Se intake of 0.065 mg/d at an energy level of 10 MJ). The increase in the amount of Se in fertilizers also increased the daily Se intake, reaching 0.08 mg/d in 2001 (Fig. 10). This is slightly higher than in the 1999 figure but probably represents a plateau level.

The most important Se sources are milk, meat, and milk and meat products. Together they account for nearly 70% of the total Se intake. An individual's Se intake may be low as a result of vegan diets which contain no milk or any other foods of animal origin. Extremely high Se intakes, exceeding the No-Observed-Adverse-Effect Level (NOAEL) of 0.4 mg/d of daily recommended intake (DRI), are probably not due to dietary sources if they are sustained over lengthy periods (Food and Nutrition Board and Institute of Medicine 2000).

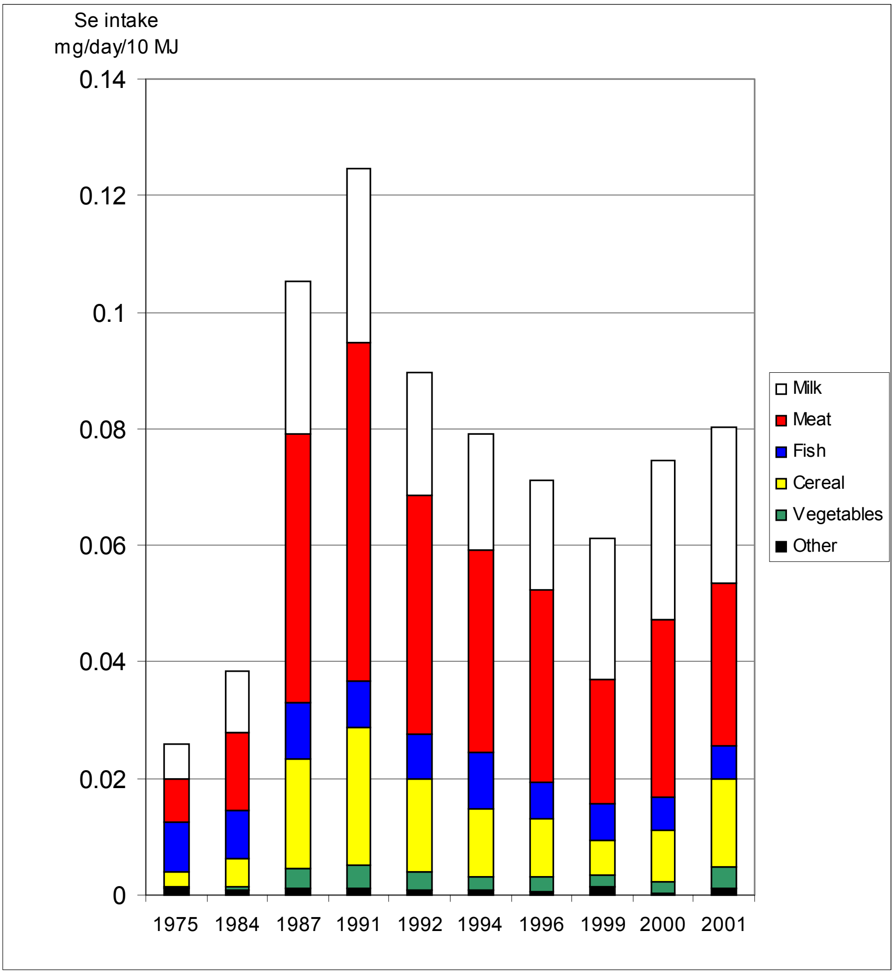


Fig. 10. Trends in Se intake in Finland during 1975-2001.

In Finland the average daily Se intake at an energy level of 10 MJ is now higher than in most other European countries, and almost at the same level as in the adequate Se areas of the USA and Canada (Food and Nutrition Board and Institute of Medicine 2000). It satisfies god the RDA and DRI Se intake recommendations of 0.055 mg/d (Food and Nutrition Board 1989, Food and Nutrition Board and Institute of Medicine 2000). The average daily Se intake is now at a very adequate and safe level in Finland.

8 Serum and whole blood selenium

8.1 Serum selenium

Serum selenium concentrations in healthy Finnish adults have been monitored regularly since the 1970s. During the 1970s, the low dietary intake of Se, 25 µg/d (Mutanen & Koivistoinen 1983), was reflected in a low serum Se level of 0.63-0.76 µmol/l (Alfthan 1988). This was among the lowest values reported in the world (Alfthan & Neve 1996). Before 1985, the mean serum Se concentration varied between 0.75 µmol/l and 1.23 µmol/, depending on the amount of imported high-selenium wheat (Alfthan 1988). Since 1985, the serum Se concentration of the same healthy male and female groups, in urban Helsinki (n=30-35) and rural Leppävirta (n=35-45), have been monitored systematically. The combined annual mean serum Se concentrations for 1984 to 2002 are shown in Fig. 11. One year before Se supplementation of fertilizers was started, the mean serum Se concentration was 0.89 µmol/l and, it reached its highest level four years later at 1.5 µmol/l, one of the highest values in Europe. After the decrease in the amount of fertilizer Se in 1990 and 1998, serum Se decreased to a new stable level of 1.10 µmol/l in 1999. This serum Se level is still above the mean value for Europe, but lower than is generally found in Canada or the USA (Alfthan & Neve 1996). Since the second change in the amount of fertilizer Se in 1998, an increasing trend has been observed.

Serum Se concentrations in subpopulations thought to be at risk of suboptimal Se intake have also been studied (Varo et al. 1994). The mean serum Se concentrations of mothers at delivery and their neonates during 1983 to 1996 have paralleled adult serum Se levels, although at a lower level. Neither exceptionally low nor exceptionally high individual values have been observed during this period.

Since 1985, whole blood Se has been monitored systematically only in the rural group. The relative effect (maximum/baseline) of Se supplementation on whole blood Se was larger than on serum Se, an increase of 125% and 70%, respectively, since 1985 (Fig. 12). Whole blood Se reached its peak approximately one year later than serum Se. Before supplementation of fertilizers with Se the mean value was 1.15 µmol/l, and at maximum the value was 2.62 µmol/l (in 1990). After 1992, the decrease paralleled that of serum Se, leveling off at 1.85 µmol/l by 1999. As a consequence of the second change in the amount of fertilizer Se mean whole blood Se has again started to increase.

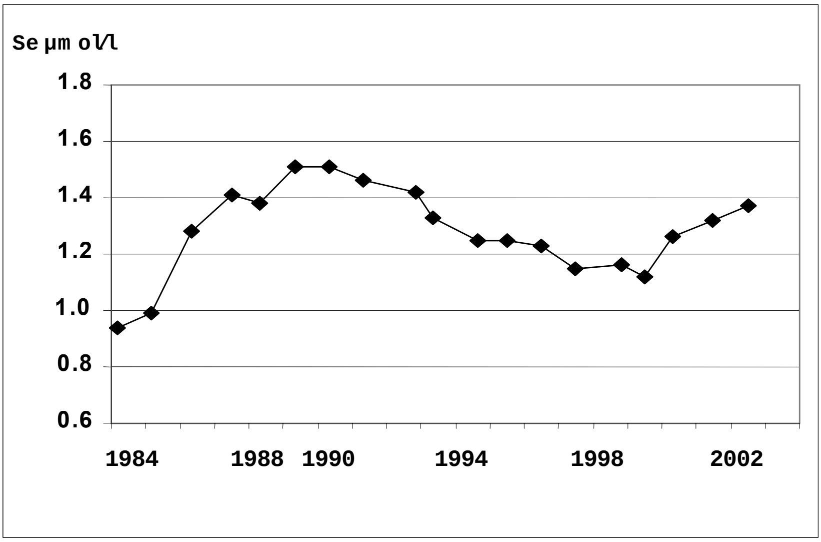


Fig. 11. Annual mean serum Se concentration in healthy Finns before and during Se fertilization

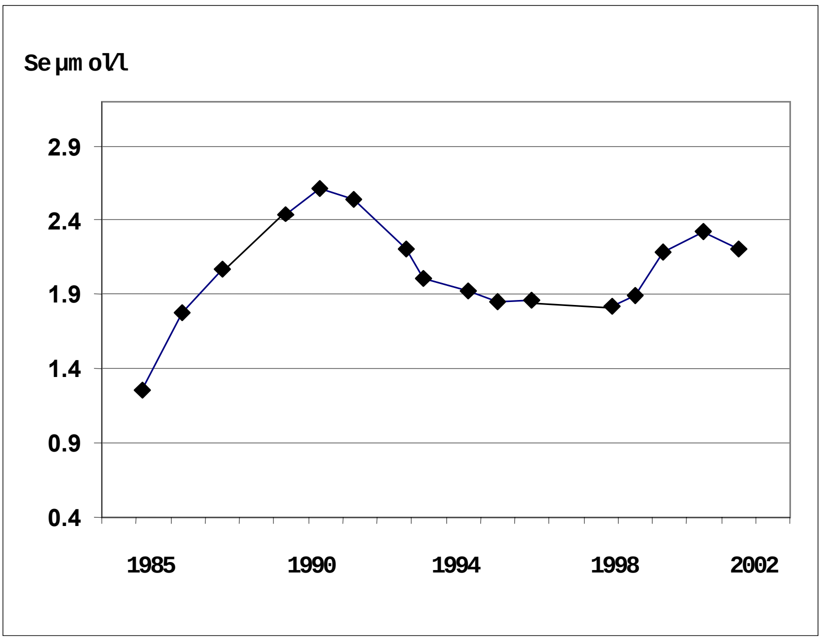


Fig. 12. Annual mean whole blood Se concentration in healthy Finns before and during Se fertilization.

8.2 Tissue selenium

Toenails reflect the integrated intake of Se over a period of 6 months to a year (Longnecker et al. 1993). Se concentrations in toenails have not been studied systematically on an annual basis. Toenail data for the period 1984 to 1995 has been compiled from different Finnish studies on groups of healthy subjects and from data on follow-up groups (Alfthan et al. 1991a, Ovaskainen et al. 1993). Before supplementation of fertilizers with Se started, the mean toenail Se concentration was 0.45 mg/kg. The maximum level, 0.91 mg/kg, was observed in 1992 about two years later than for whole blood Se. In accordance with serum and whole blood Se, a clear downward trend was seen after the first change in the amount of fertilizer Se and the latest value, from 1995, is 0.72 mg/kg. The inter-individual variation in toenail Se was typically 10-12%. In a European multicenter study comprising eight countries, the toenail Se concentration of 59 middle-aged Finnish men sampled during 1990-1992 was the highest, 0.84 mg/kg (Virtanen et al. 1996).

Liver Se accounts for the largest fraction of body stores of Se. Its Se is mobile and reflects dietary Se intake over a time span of weeks (Levander et al. 1983a). Se has been determined in human liver samples obtained at autopsy from men who had died in traffic accidents both before (1983-1985) and during (1988-1989) the period of Se supplementation of fertilizers. Initially, the mean value was 0.95 ± 0.27 mg/kg dw (n=53). The increased intake of Se increased the mean Se concentration of liver tissue by 60% (to 1.58 mg/kg) in samples (n=12) obtained 3-4 years later (Varo et al. 1994). This value was slightly low compared with data obtained from other European countries.

8.3 Serum, red blood cell and platelet glutathione peroxidase activity

The activity of the Se-dependent enzyme, glutathione peroxidase (GSHPx) is associated with Se intake only at moderate intake levels. At higher Se intakes, the activity of the enzyme in serum and whole blood reaches a plateau and cannot be stimulated further. The plateau, in terms of serum Se, is under 50 µg/d, and for whole blood less than approximately 60-80 µg/d (Yang et al. 1987, Levander 1989, Alfthan et al. 1991b). Saturation of serum GSHPx activity has been regarded as a measure of optimal Se intake and has been the basis of the current US Recommended Dietary Allowance (Levander 1989).

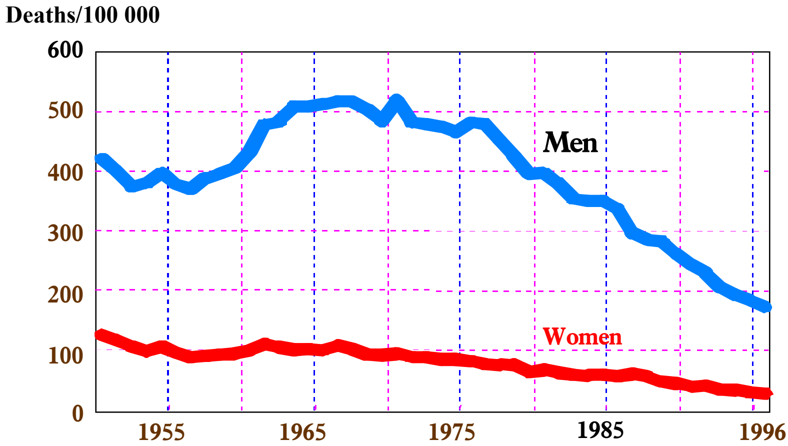
Two placebo-controlled supplementation studies have been carried out in Central Finland on the same 50 middle-aged healthy male blood donors (Levander et al. 1983b, Alfthan et al. 1991a). The aim of the studies was to find out to what extent platelet GSHPx activity could be increased by Se supplementation and the qualitative effect of organic/inorganic Se supplementation on GSHPx activity. The first study was performed in 1981 (supplementation for 11 weeks) and the second in 1987, that is to say before and during Se fertilization (supplementation for 16 weeks). In both studies, 10 men were supplemented with 200 µg of organic Se in the form of Se-enriched yeast or with 200 µg sodium selenate. The third group received a placebo. At baseline in 1981, the mean plasma Se concentration was 0.89 µmol/l and in 1987 1.40 µmol/l, which reflected mean dietary Se intakes of 39 µg/d and 100 µg/d, respectively.

The percentage increase in platelet GSHPx activity for men supplemented with either selenate or yeast Se was calculated in relation to the activity of the placebo groups. Before the addition of Se to fertilizers, selenate and yeast-Se supplements increased the enzyme activity by 104% and 75%, respectively. During fertilization the effects of selenate and yeast-Se were much lower, 41% and 6%, respectively. The results suggested that an intake of 100 µg Se per day was still not sufficient to completely saturate GSHPx activity in platelets. At the higher intake level, however, both plasma and red blood cell GSHPx activities were maximally stimulated. Extrapolation of platelet data, including the two Finnish studies and six other studies with a similar design, suggests that maximal stimulation of platelet GSHPx activity would occur at a plasma Se level of 1.3 µmol/l (Alfthan et al. 2000).

9 Human health

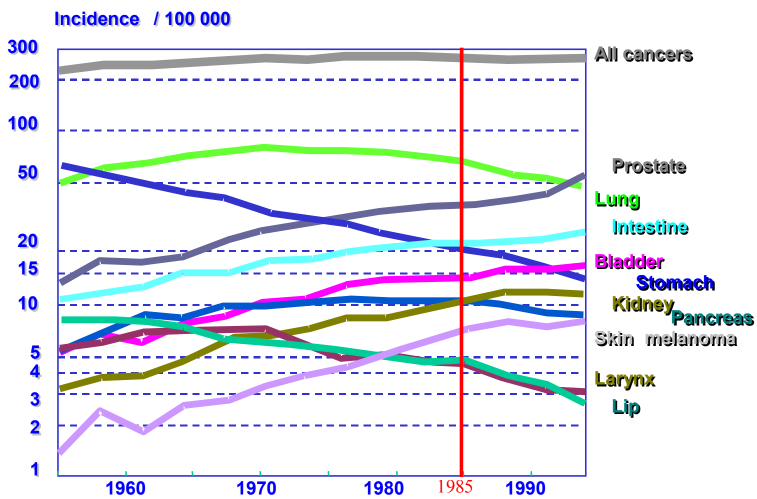
The supplementation of fertilizers in Finland is a nationwide experiment affecting all individuals, not a placebo-controlled clinical trial. Therefore it is impossible to measure in exact terms the health outcomes of the intervention. Apart from Keshan disease, which can be prevented by prophylactic administration of sodium selenite (Keshan Disease Research Group 1979), the role of Se in human disease in unclear. Judging from the results of epidemiological studies, mortality and morbidity in cardiovascular diseases (Salonen et al. 1982) and cancer (Salonen et al. 1984, Knekt et al. 1990) are endpoints that might have been affected by increasing the Se intake in Finnish people. If it is anticipated that Se exerts its effects via the antioxidant activity of the Se-dependent enzyme, GSHPx, then the intervention should have been successful, since it resulted in near-maximal stimulation of GSHPx activity in all blood components (Alfthan et al. 1991a).

Age-adjusted mortality from coronary heart disease has declined continuously in Finland since the end of the 1960s (Fig. 13). Most of the decline can be attributed to favourable changes in the levels of classical risk factors such as serum cholesterol, smoking and blood pressure and, since 1985, it has been impossible to find any change in the declining trend that could be attributed to Se supplementation (Pietinen et al. 1996). Neither do the data on cancer incidence in Finland suggest any specific effects due to increased dietary Se intake in the late 1980s (Fig. 14). For example, lung cancer incidence



Source: Statistics Finland

Fig. 13. Age-standardized mortality from coronary heart disease in Finland during 1955-1996. Deaths per 100,000 of the population aged 35-64



Source: Finnish Cancer Registry

Fig. 14. Age-standardized incidence from cancers in males in Finland during 1955-1995.

in males shows a downward trend from the mid 1970s to 1985, with no change in the rate thereafter (Fig. 15). These diseases are affected by numerous different factors and it will be extremely difficult to determine whether the increased Se intake resulting from supplementation of fertilizers has influenced the health of the nation. The most important reason for this is the

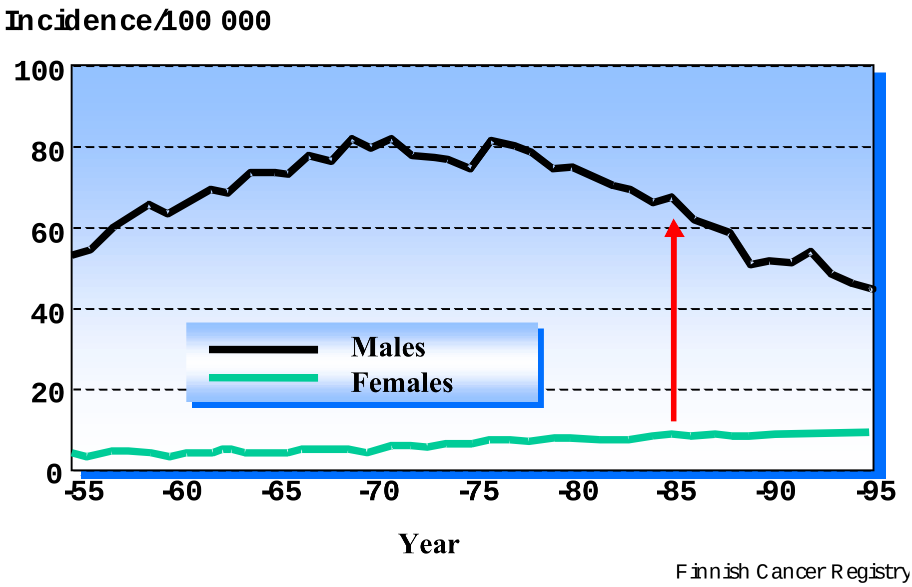


Fig. 15. Age-standardized incidence of lung cancer in Finnish males and females.

lack of a reference population, since the entire population of Finland (5.1 million) is affected equally by the increased Se intake. Cultural, dietary and genetic differences between Finnish and neighboring populations prevent any meaningful comparison. Conclusions on the effects of increased Se intakes should be based on controlled clinicat trials.

10 Environment

No health hazards from possible environmental effects of Se supplementation have been observed to date. Studies on the Se levels of tap water, groundwater, lake and river waters and lake sediments from samples taken during 1990-1992 have disclosed no environmental effects that could be ascribed to the supplemented fertilization (Alfthan et al. 1995, Wang et al. 1991; 1994; 1995). A follow-up study was done in 1997-1999 on the seasonal variation of water Se from 14 rivers and lake sediments from seven lakes (Eurola & Hietaniemi 2000). During 1997, the mean Se concentration of river waters was lowest in June (92 ng/l) and highest in August (119 ng/l). The mean values were similar to those measured in 1990 and 1991, at 104 and 123 ng/l, respectively. In Table 8, the results for both water and sediment Se concentrations are shown for samples (1 cm layers from top to bottom, 1-30 cm) taken in 1992 and 1999. The mean water Se concentration did not differ between the two sampling years. The mean selenium concentrations of the sediment top layers sampled in 1999 were only slightly lower than in 1992. In five of the lakes, the Se concentration in sediments was higher in the top layers than the bottom layers, approximately corresponding to the time after and before fertilization started, but an increase Se concentration was already seen during the first half of the 1900s (Wang et al. 1995). The amounts of Se that have been used annually in fertilizers, 20 tons during 1985-1990 and 7.6 tons during 1991-1998, are comparable with the total fallout of Se from precipitation, estimated to be 18 tons in 1989 (Wang et al. 1993).

Table 8: Lakewater and sediment Se in 19th century.

Lake	Thropic level	Water 1999 Se ng/l	Water 1992 Se ng/l	Sediment Bottom 1999 Se mg/kg dw	Sediment Top 1999 Se mg/kg dw	Sediment Top 1992 Se mg/kg dw
Pyhäjärvi	+	115	81	0.18	0.23	0.23
Villikkalanjärvi	+	162	113	0.23	0.16	0.27
Kyöliönvesi	+	116	59	0.31	0.45	0.35
Onkivesi	+	91	58	0.26	0.37	0.26
Pääjärvi	±	99	143	0.71	0.49	1.05
Oso-Hietajärvi	-	40	34	1.16	2.06	2.03
Pesosjärvi	-	52	89	2.95	2.82	3.64
Mean		96	82	0.82	0.94	1.12

Bottom sediment 1999 is mean sediment layers below representing 19th century

11 Summary and conclusions

For climatic and geochemical reasons, Finland is a low-Se area. On the basis of studies of the Se content of foods, epidemiological associations, and field trials of Se supplementation, it was decided in 1984 to increase the Se content of foods and the Se intake of the population by supplementing compound fertilizers with Se in the form of sodium selenate. Within two years, a three-fold increase in Se intake was observed to levels of 100-120 μg per day in 1989-1991. The supplementation affected the Se content of all major food groups with the exception of fish. Serum and whole blood Se concentrations of people increased concomitantly by 70% and 125%, respectively. In 1990, the amount of Se added to fertilizers for grain production was reduced by about 60%. This reduced the Se intake by 30% and mean serum and whole blood Se levels by 20% and 40%, respectively, from the highest levels observed in 1989 and 1991. Plants take up part of the supplemented selenate and convert it into organic Se compounds, mainly selenomethionine. This affects human nutrition by increasing the Se content of foods of both animal and vegetable origin. It also reduces the needs to supplement animal feeds with inorganic Se.

In Finland, where the geochemical conditions are relatively uniform, the supplementation of fertilizers with Se has proved to be an effective, safe and controlled way of bringing the Se intake of the whole population of to the recommended level (Ekholm 1997). Since most of the supplemented Se is derived from foods of animal origin, supplementation of animal feeds with organic Se would be an alternative way of increasing the Se concentration in foods and the human Se intake (Aro 1996).

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