

# Effects of Selenium Fertilization on the Selenium Content of Cereal Grains, Flour, and Bread Produced in Finland

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## ABSTRACT

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In order to raise the selenium (Se) content of agricultural products in Finland, the main multinutrient fertilizers have been supplemented with sodium selenate since the fall of 1984. This intervention has affected most locally produced agricultural products. The Se contents of grain (spring and winter wheat and rye), flour, and bread have been monitored regularly using electrothermal atomic absorption spectrophotometry every other month since 1983. The supplemented fertilization has distinctly raised the Se contents of cereals and cereal products, spring wheat showing the greatest increase (20–30-fold). The mean Se values (mg/kg dry matter) in 1986–1988 were: spring wheat 0.25, winter wheat 0.05, rye 0.04, wheat flour 0.17, rye flour 0.07, wheat bread 0.18, rye bread 0.04, and rolled

oats 0.19. Before 1985 the level of Se in all domestic cereal grains was 0.01 mg or less per kilogram. The Se content of flours is also affected by the proportion of imported to domestic grain in milling. In the 1980s, the amount of imported grain has varied from 0 to 65%. Much of the grain has been imported from North America, where grain usually has a high Se content; the range in the present study was 0.03–1.50 mg/kg dry matter. At present, American grain raises the Se content of Finnish cereal products, whereas other imported grain reduces it. The average Se intake in Finland is now about 0.11 mg/day per capita at the energy level of 10 MJ (2,400 kcal), with cereals contributing about 18% of the total intake.

For geochemical and climatic reasons, selenium (Se) available from the soil in Finland has been exceptionally low (Koljonen 1975, Sippola 1979). Consequently, the Se contents of domestic agricultural products have also been low (Koivistoinen 1980). In the 1970s, the estimated average Se intake in Finland was about 0.02–0.03 mg/day (Varo and Koivistoinen 1980, Mutanen 1984), which was well below the safe, adequate intake level for adults (0.05–0.2 mg/day) defined by the U.S. National Academy of Sciences' National Research Council (1980). The low intake level was considered to be a potential nutritional risk for the Finnish population (Huttunen and Koivistoinen 1985).

Cereals are normally an important source of dietary Se, but the Se content of cereal grains grown in Finland has been only about 0.01 mg/kg dry matter or less (Oksanen and Sandholm 1970, Varo et al 1980). In normal years, 5–10% imported grain is mixed with domestic grain in milling. In poor harvest years, the proportion of imported grain may be greater, even more than 50%. The Se content of grain imported from North America is usually high. Consequently, the Se content of cereal products and the average Se intake has been highly dependent on the ratio of domestic to imported cereal grains in milling. For example, the Se intake was doubled by the abundant grain imports in 1979–1980 (Varo and Koivistoinen 1981).

In order to raise the Se content of domestic agricultural products, and also to increase and stabilize the Se intake, an official decision was made to supplement the main agricultural multinutrient fertilizers with sodium selenate starting in the fall of 1984 (Ministry of Agriculture and Forestry 1984). Because selenate is transported more efficiently to the vegetative parts of the plant than to the grain (Ylärinta 1985), two supplementation doses are used: 16 mg Se/kg in fertilizers mainly used in grain production, and 6 mg/kg in fertilizers used in hay and fodder production. The amount of Se introduced by fertilization is about 8 g/hectare (ha) for cereal crops and 3 g/ha for grass crops (Ylärinta 1985). The primary objective of this intervention was to raise the Se level of domestic cereal grains to an approximate level of 0.1 mg/kg of dry matter. However, it was expected that other agricultural products would also be affected. An expert group was appointed by the Ministry of Agriculture and Forestry to evaluate and report annually the effects of the Se fertilization. The Se concentration of fertilizers, soils, animal feeds, human

sera, and 13 basic foods are monitored regularly. Occasionally other selected foodstuffs also have been analyzed.

The present paper describes the effects of Se fertilization on the Se content of cereal grains and its consequences on Se intake in Finland. The effects of cereal grain imports in the present situation are also discussed.

## MATERIALS AND METHODS

### Samples

The samples of bread grain cereals (spring and winter wheat and rye) were received once a year from major commercial mills and from State Granary silos located throughout Finland. Each sample (1–5 kg) represented about 0.01–1.5 million kilograms of grain. The sampling system was considered to give a good overview of the agricultural situation throughout the country. The samples of imported grain were received from State Granary storages, each sample representing one imported batch of grain.

Se levels of wheat and rye flours and breads are sampled systematically six times annually. The flour samples are taken straight from the production line of the six major commercial mills, one rye and one wheat flour sample from each mill per sampling period, giving 36 samples annually. In practice, the number of samples received from the mills was somewhat lower. The bread samples are purchased from retail food stores in the Helsinki area, eight samples per sampling period. Original samples were pooled in pairs to make four analyzable samples—24 samples per item annually.

### Pretreatment

Grains were ground to make the samples more homogeneous. All samples were packed into polyethylene bags, about 200 g into each bag, and stored at  $-20^{\circ}\text{C}$  until analysis (two to four weeks).

### Analytical Method

Se was analyzed by an electrothermal atomic absorption spectrophotometry method for food samples described previously (Kumpulainen et al 1983). The dried samples were digested in a mixture of concentrated  $\text{HNO}_3$ ,  $\text{HClO}_4$ , and  $\text{H}_2\text{SO}_4$ . Se was reduced to Se(IV), chelated with ammonium pyrrolidine dithiocarbamate, and extracted into methylisobutylketone for the spectrophotometric determination.

The accuracy of the method was tested by determining three certified reference materials. An unofficial control material was analyzed continuously as a blind sample to test the precision of the analytical method (Table I).

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## RESULTS

The Se levels of wheat and rye before and after the beginning of the Se fertilization are given in Table II. The effect of selenate supplemented fertilization on cereal grains is very distinct. The Se concentration of spring wheat increased 20–30-fold. In winter cereals, Se levels increased two- to fivefold. Spring wheat corresponds to about 95% and winter wheat to about 5% of the total wheat production in Finland (State Granaries 1989).

Se fertilization also affected the Se contents of flour and bread (Table III, Fig. 1). The influence on Se levels of rye flour and rye bread has been slight, whereas the Se contents of wheat flour, wheat bread, and rolled oats increased 10–20-fold.

In addition to the Se from fertilizers, the Se content of flour is also affected by the proportion of imported grain in milling. In the 1980s, the proportion of imported wheat varied from 0 to 65%, and that of imported rye from 20 to 65% (Table IV). The origin and the Se content of imported grain in 1981–1988 are presented in Table V. Most of the imported wheat has been North American and its Se content has been generally high. The

origin of the imported rye has been more variable.

The Se-supplemented fertilization has affected Finnish agricultural products of both plant and animal origin. Hence, the total Se intake has risen from 0.025 mg/day in the mid-1970s to about 0.11 mg/day per capita during 1985–1988 (Fig. 2) (Ministry of Agriculture and Forestry 1989). At present, cereals contribute about 18% of the estimated average Se intake in Finland, and their relative significance as a source of Se has doubled since the 1970s. Most of the Se is obtained from wheat products, as both the consumption and Se content of wheat are higher than those of rye. The annual consumption of wheat is approximately 44 kg, but that of rye 19 kg per capita.

## DISCUSSION

The selenate supplementation of fertilizers has been effective in raising the Se content of cereal grains in Finland. The effects of Se fertilization became evident in the growing season of 1985, as soon as selenate-supplemented fertilizers were in common use. The Se content of spring wheat has now stabilized at an average level of 0.25–0.30 mg/kg of dry matter, which is higher than the original minimum target level (0.1 mg/kg of dry matter). The maximum Se content encountered in domestic spring wheat samples has been 0.47 mg/kg dry matter.

The increment in the Se content of rolled oats is also clear. The transport of Se from soil to grain seems to be similar in oats and spring wheat. Both are spring cereals, and consequently their fertilization practices are also comparable.

The Se level of winter cereals has remained considerably lower than that of spring wheat, the average Se level of rye and winter wheat now being about 0.04 mg/kg of dry matter. The reason for this may be differences in fertilizing and cultivating practices. In fall, during sowing, winter cereals are treated with only a light dose of Se-supplemented fertilizer, whereas in spring, plain nitrogen fertilizer without Se is applied more generously. Thus the amount of Se introduced to winter cereals is smaller than that given to spring cereals. However, some of the selenate may be leached during wet fall conditions. Also, selenate may be reduced to selenite during the winter. Selenite and other reduced forms of Se are readily bound to soil constituents, and are thus unavailable to plants in the growing season (Cary and Allaway 1969, Cary and Gissel-Nielsen 1973, Hamdy and Gissel-Nielsen 1977). Strong immobilization of selenite may be the main reason for the negligible accumulative effect of Se applications.

The trends in Se contents of wheat and rye flour and wheat and rye bread are seen in Figure 1. Since the fall of 1985, the Se contents of wheat flour and bread have risen markedly. In these products, the Se level has generally been about two-thirds of that in domestic grain. Toward the end of 1988, the Se contents of flour and bread were increased sharply by the higher proportion (25%) of North American grain in milling. The Se content of rye products also increased, but the changes have been slow.

The Se content of a cereal plant reflects the soluble Se status of the soil. For geochemical reasons, North American wheat and rye are usually higher in Se than wheat and rye grown in other parts of the world (Morris and Levander 1970, Arthur 1972, Olson and Palmer 1984, Fransson 1987). Most of the wheat imported to Finland has been North American, and the annual averages

**TABLE I**  
Accuracy and Precision of the Analytical Method  
(Se content mg/kg dry matter)

Sample	n	Present Study	Certified Value
		Mean ± SD	Mean ± SDC
NBS 1577a (bovine liver)	6	0.66 ± 0.03	0.71 ± 0.07
NBS 1569 (Brewer's yeast)	3	0.92 ± 0.03	0.92 ± 0.09 <sup>a</sup>
IAEA H-9 (human diet)	2	0.10	0.11
Noncertified control <sup>b</sup>			
Rye flour	62	0.028 ± 0.003	

<sup>a</sup> Not certified by NBS (Gladney 1980).

<sup>b</sup> Analyzed as blind samples.

**TABLE II**  
The Se Content of Finnish Wheat and Rye in 1982–1988  
(mg/kg dry matter)

Sample	n	Mean ± SD	Range
1982–1984			
Wheat	30	0.01 ± 0.01	<0.01–0.03
Rye	26	0.01 ± 0.01	<0.01–0.02
1985			
Spring wheat	17	0.23 ± 0.10	0.08–0.40
Winter wheat	4	0.03 ± 0.02	0.01–0.05
Rye	9	0.02 ± 0.02	<0.01–0.07
1986			
Spring wheat	23	0.23 ± 0.06	0.13–0.35
Winter wheat	15	0.05 ± 0.03	0.02–0.13
Rye	27	0.04 ± 0.02	0.02–0.11
1987			
Spring wheat	11	0.27 ± 0.11	0.11–0.47
Winter wheat	7	0.04 ± 0.04	0.02–0.12
Rye	12	0.03 ± 0.01	0.01–0.05
1988			
Spring wheat	15	0.29 ± 0.07	0.20–0.39
Winter wheat	7	0.04 ± 0.03	0.01–0.09
Rye	7	0.04 ± 0.02	0.02–0.07

**TABLE III**  
Se Levels of Flour, Bread, and Rolled Oats (mg/kg dry matter) Before and After Beginning Se-Supplemented Fertilization (in 1985)

Sample	Before Se Supplementation						After Se Supplementation					
	1975–1976			1983			1986			1988		
	n	$\bar{x} \pm SD$	Range	n	$\bar{x} \pm SD$	Range	n	$\bar{x} \pm SD$	Range	n	$\bar{x} \pm SD$	Range
Wheat flour (0.6% ash)	9	0.02 ± 0.01	<0.01–0.02	29	0.04 ± 0.02	0.01–0.07	29	0.16 ± 0.04	0.07–0.23	35	0.19 ± 0.07	0.03–0.30
Rye flour (whole grain)	10	0.01 ± 0.01	<0.01–0.03	29	0.07 ± 0.05	0.01–0.23	29	0.02 ± 0.01	0.01–0.06	35	0.06 ± 0.06	0.01–0.29
Wheat bread (French bread)	5	0.01 ± 0.00	<0.01–0.02	24	0.05 ± 0.03	0.01–0.11	24	0.16 ± 0.03	0.11–0.21	24	0.20 ± 0.05	0.18–0.30
Rye bread	7	0.02 ± 0.02	0.01–0.05	24	0.08 ± 0.07	0.01–0.27	24	0.03 ± 0.02	0.01–0.09	24	0.05 ± 0.02	0.02–0.09
Rolled oats	8	0.01 ± 0.01	<0.01–0.02				4	0.19 ± 0.06	0.14–0.27			

<sup>a</sup> Varo et al 1980.

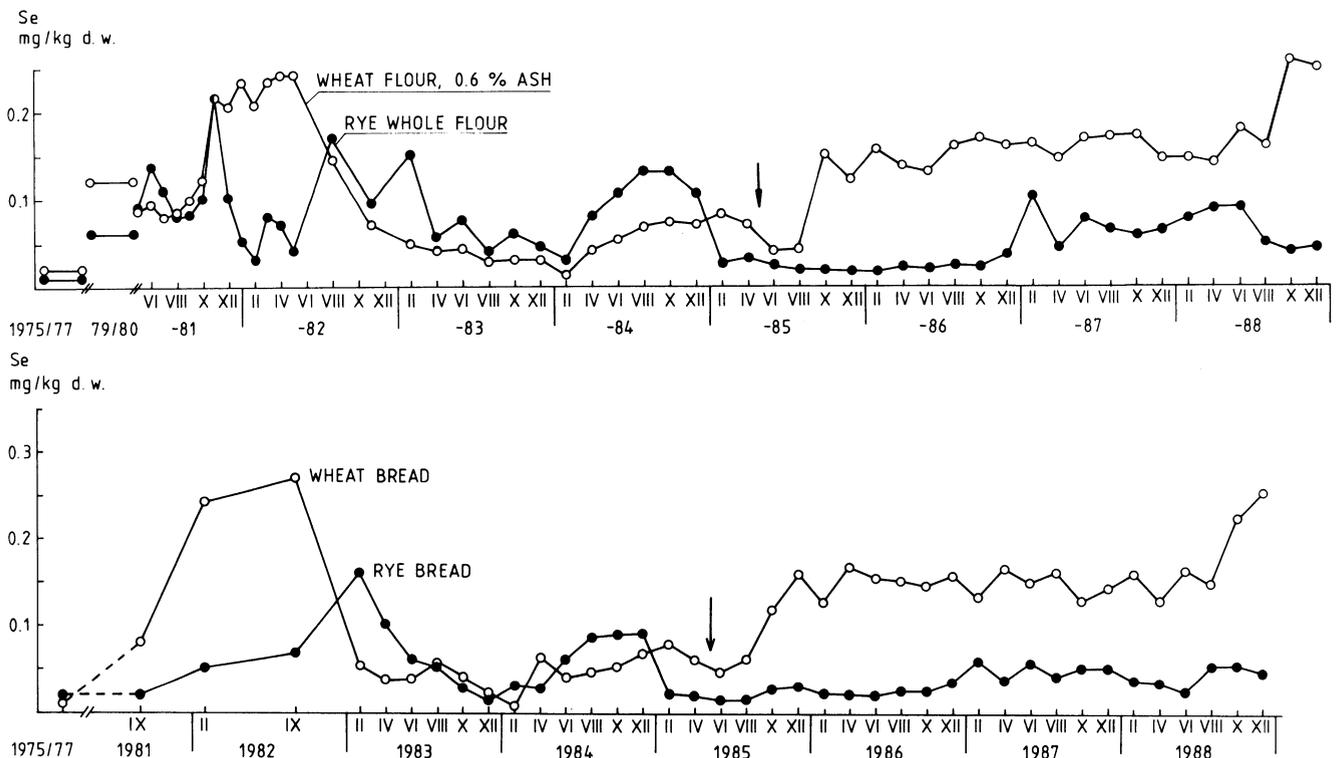


Fig. 1. Trend in the selenium contents of rye flour, rye bread, wheat flour, and wheat bread. The arrow in the figure marks the introduction of Se-supplemented fertilization. Se values for 1979–1980 are from Varo and Koivistoinen (1981).

TABLE IV  
Percentages of Domestic Wheat and Rye Used in Milling in 1980–1988

Grain	Data Collection Period	%
Wheat	Jan. 1, 1980 to Apr. 30, 1980	50
	May 1, 1980 to Aug. 31, 1980	40
	Sept. 1, 1980 to Sept. 30, 1981	80
	Jan. 1, 1981 to Oct. 31, 1981	70
	Nov. 1, 1981 to Sept. 14, 1982	35
	Sept. 15, 1982 to Oct. 30, 1982	70
	Sept. 1, 1983 to Feb. 29, 1984	100
	Mar. 1, 1984 to May 31, 1985	100 <sup>a</sup>
	June 1, 1985 to Oct. 31, 1985	85
	Nov. 1, 1985 to Jan. 31, 1988	95
Rye	Feb. 1, 1988 to Sept. 30, 1988	85
	Oct. 1, 1988 to Dec. 31, 1988	75
	Jan. 1, 1980 to Feb. 28, 1980	50
	Mar. 1, 1980 to Aug. 31, 1980	60
	Sept. 1, 1980 to Sept. 30, 1981	80
	Jan. 1, 1981 to Oct. 31, 1981	70
	Nov. 1, 1981 to Dec. 31, 1982	35
	Jan. 1, 1983 to July 31, 1983	47
Sept. 1, 1983 to June 30, 1988	70 <sup>b</sup>	
Feb. 1, 1988 to Sept. 30, 1988	70 <sup>b</sup>	
Oct. 1, 1988 to Dec. 31, 1988	65	

<sup>a</sup>Mills were allowed to use 15% imported wheat provided they had exchanged the equivalent amount of domestic wheat with the State Granary.

<sup>b</sup>Since April 1987, mills have been allowed to use, at most, 10% imported rye, which is the same price as domestic rye. Mills must use not less than 60% domestic rye.

in Se content have varied between 0.3 and 0.6 mg/kg dry matter, the maximum single value being 1.5 mg/kg. North American rye has also generally been high in Se.

At present, the Se level in Finnish spring wheat is approximately half of that common in North American wheats, but it is clearly higher than in European and other wheats.

The effect of grain imports is evident in Figure 1. The Se contents of wheat flour and wheat bread were especially high in the early 1980s and at the end of 1988. In 1981–1982 the proportions of

TABLE V  
Origin and Se Content of Cereal Grains Imported to Finland (mg/kg dry matter).

Grain	<i>n</i>	Mean	Range
<b>Wheat</b>			
1981–1982			
North America	5	0.43	0.11–0.94
1984			
North America	6	0.32	0.03–0.50
1986			
North America	3	0.33	0.05–0.58
Sweden	1	0.02	...
1988			
North America	11	0.56	0.04–1.50
Saudi Arabia	1	0.12	...
Hungary	2	0.05	0.05–0.05
<b>Rye</b>			
1981–1982			
North America	3	0.30	0.05–0.45
Denmark	3	0.03	0.01–0.05
Germany	3	0.04	0.02–0.08
Sweden	1	0.02	...
1984			
Denmark	2	0.14	0.03–0.25
Sweden	1	0.02	...
Poland	1	<0.01	...
1986			
North America	2	0.36	0.34–0.38
Denmark	1	0.02	...
Germany	2	0.02	0.01–0.02
Sweden	1	0.02	...
Poland	1	0.02	...
1988			
Germany	6	0.02	0.01–0.02
Soviet Union	3	0.03	0.02–0.04
Hungary	1	0.03	...

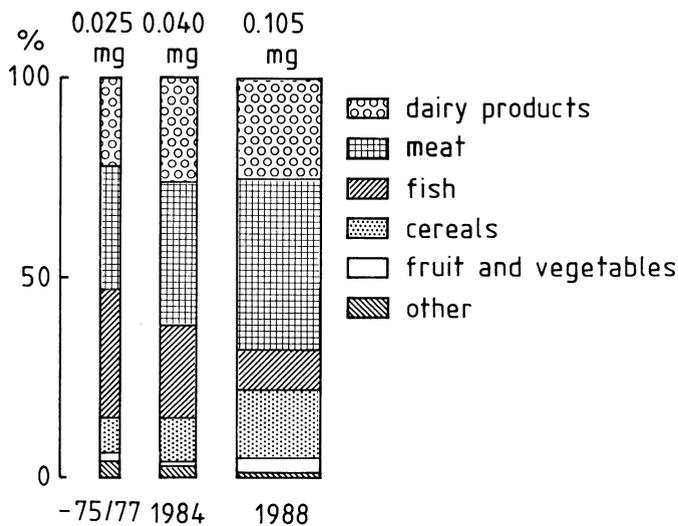


Fig. 2. The total average selenium intake ( $\mu\text{g}/\text{day}$ ) in Finland in 1975–1976, 1984, and 1988 at a consumption energy level of 10 MJ/day.

imported wheat and rye in milling were temporarily raised to 65%. The imported grain was mainly North American (Table V). In October 1988, the proportion of imported wheat in milling was raised from 5 to 25%, the origin of the imported wheat again being North America. However, the rye imported in 1988 was mainly European. Thus the Se levels of rye flour and bread remained at a moderate level, although the proportion of imported rye in milling was 20–25%.

Se-supplemented fertilization has raised the Finnish population's estimated average Se intake to about 0.11 mg/day at an energy level of 10 MJ (2,400 kcal). It is now well within the limits of safe and adequate intake for adults, 0.05–0.20 mg/day (National Research Council 1980), and it somewhat exceeds the proposed Nordic recommendation, 0.04–0.08 mg/10 MJ (Nordisk Ministerråd 1989). Cereals are the most important plant source of Se, contributing about 18% of the total Se intake in Finland. In the present situation, large grain imports from North America would increase the average Se intake; imports from most other countries would have the opposite effect.

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