

# Cropland Water-Soluble Selenium, Groundwater Silicon, Atlantic Rain – Agro-Geological Assessments

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## ABSTRACT

Soil selenium (Se) is known to be associated with its clay content and organic matrix. The role of the water-soluble Se [Se.H<sub>2</sub>O], “plant available Se”, and its atmospheric Se supply needs clarification. The main aim is to compare regional associations of [Se.H<sub>2</sub>O] and groundwater silicon [Si] with erosion associated Latitude [Lat], longitude [Long] and Altitude [Alt], as well as with Atlantic rain {determined by its change in oxygen isotope ratio (<sup>18</sup>O/<sup>16</sup>O) from its Atlantic value: [δ180]}, groundwater hardness [Ca+Mg] and [Mg+K] (as biotite/clay indicator) in order to find differences in their terrestrial and atmospheric associations.

**Results:** [Si] was better explained by [Lat], [Long], [Alt] and [δ180] than by [Se.H<sub>2</sub>O]. [Ca+Mg] and [Mg+K] associated stronger with [Se.H<sub>2</sub>O] (p < 0.001, by both) than with [Si] (p < 0.01, by both). Combined regressions by [Lat;Long;Alt] explained [Se.H<sub>2</sub>O], [Si] and [δ180] by 65.2 – 95.2 % (p < 0.001, by all), all coefficients were strongly negative by [Si.gw], but coefficients of [Alt] were positive by [Se.H<sub>2</sub>O] and [δ180]. [Si] explained [Se.H<sub>2</sub>O] by 61.9 % (p < 0.001).

**Conclusions:** [Si.gw] was more strongly than [Se.H<sub>2</sub>O] associated with geographic factors, but [Se.H<sub>2</sub>O] with clay-indicator [Mg+K], as well as with [Ca+Mg]. Positive coefficient of [Se.H<sub>2</sub>O] with [Alt] in combined regression by [Lat;Long;Alt] suggests “hint-like” that the altitude-erosion of Se could have been compensated by atmospheric Se. High association between [Si] and [Se.H<sub>2</sub>O] can be explained by their associations with humus. Se fertilization seemed to have influence on [Se.H<sub>2</sub>O] and its ability to predict plant Se.

## Introduction

This study is treating hot water extractable selenium of Finnish croplands in 1978-80, before the era of Se fertilizers. There are many methods for cropland Se determination [1], even “total selenium” can be determined by different selections and orders of strong acids. Se values are usually expressed by mg/kg, but water-soluble Se by mg/l or µg/l. Total Se in plough layer has reported to have been 0.201 mg/kg (N=250) [2], 0.209 mg/kg (N=93) [3], 0.229 mg/kg in 1998 [4]. Generally, Se content has been highest in clays and organic soils (org), lowest in coarse mineral soils (coms). In 1979 Se content in coarse mineral soils (coms) (without silt) was 0.157 mg/kg, (calculated) [2] and in fine sandy soils 0.172 mg/kg

(even 1979) [3]; in fine mineral soils [(clay + silt), fims] 0.276 mg/kg [2] (calculated), in clays 329 mg/kg [2] and 0.290 mg/kg (early spring) [3]; in mull soils 0.228 mg/kg [2] – in peat soils 0.093 mg/kg [2] - in organogenic soils 0.464 mg/kg [3]. In studies of Ylärinta in 1983 and 13 yrs after start of Se fertilization [4], Se of organic soils was about double to that in fine mineral soils and that about 50 % higher than respective Se in coarse mineral soils. (Coms in Sippola 1979 originally included silt). Se has been associated with clays and organic matter. Clay fraction consists markedly of micas, e.g., biotite, relatively high in selenium [5]. Biotite K(Mg,Fe)<sub>3</sub>(AlSi<sub>3</sub>O<sub>10</sub>)(F,OH)<sub>2</sub>, contains Si potassium (K) and magnesium (Mg), but no calcium

(Ca) [6]. It is weatherable by normal organic acids [7]. Surprisingly amount of the liberated (molybdate reactive) Si has been only about 1 % relative to the large proportions of cations in the extracts [7]. Colloidal elemental selenium is electrically charged and adsorbed by clay minerals [5], which explains Se clay association.

Water-soluble Se can be determined by shaking or boiling the water [8], the exact method is not always clearly expressed, e.g. "soluble Se" 0.011 mg/l in [2] was extracted by acid ammonium acetate-EDTA, was not an indicator of water-soluble Se as written in [4]. Proportion of total Se: water-soluble selenium composed via hot water extraction, 3-10 % according to (Table 3) (by 7-13 $\mu\text{g}/\text{kg}$ ) in [1] and 1 % according to text in [1], 2 % (by 6  $\mu\text{g}/\text{kg}$ ), via water shaking method without heating according to the study in 13 EU countries [9], (N = 128) and ca 4 % (by 6-18  $\mu\text{g}/\text{kg}$ , in plough layer of different soil-types) by hot water extraction according to [3], N = 230. Proportion of water-soluble Se can vary from 0.3 to 36 % of total Se, according to several referates in [8]. "Soluble Se" can indicate [acid (pH 4.65) ammonium acetate-Na<sub>2</sub>EDTA (AAAc-EDTA) extractable Se] [9], which was 5 % when Se.H<sub>2</sub>O was 2 % of total selenium. "Soluble Se" can even be a synonym for acid ammonium oxalate extractate, by [(0.18 M(NH<sub>4</sub>)<sub>2</sub>(COO)<sub>2</sub>+0.1 M(COOH)<sub>2</sub>, pH 3.3]], which was 5-10 x higher to Se.H<sub>2</sub>O, Table 3 in [1]. In study of [9] the soil samples were collected from 13 European countries, samples of [1] from Finland. In the study of [9] plant Se correlated with soil selenium as follows: Water extr.  $r = 0.33^{***}$ , AAAc-EDTA extr.  $r = 0.33^{***}$ , HNO<sub>3</sub>-HClO<sub>4</sub> digest.  $r = 0.27^{**}$ , aqua regia digest.  $r = 0.23^{**}$ .

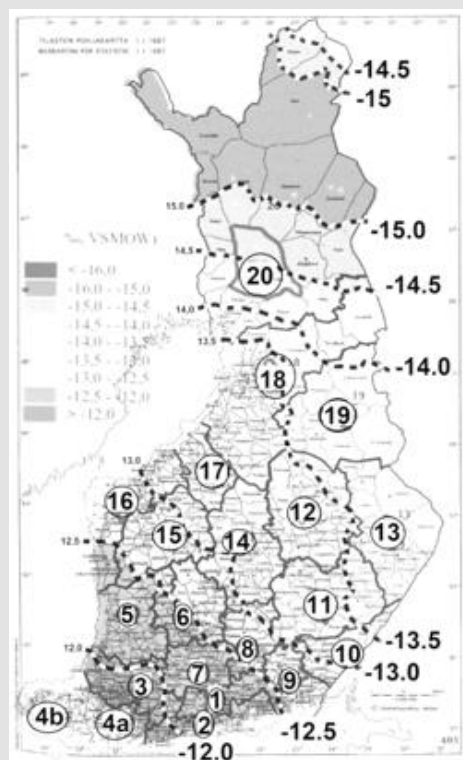
Atmospheric Se: Volatilization of selenium from selenates and selenites in Finland according to [10] for 3 months has been very scanty: generally, < 1 %, anyhow from Carex peat Se losses could reach ca 3 % by treating soils with both lime and organic matter [10]. Methylated selenium compound can volatilize more easier, e.g. even 30 % of the selenium added to fine sand in the form of trimethylselenonium chloride (Se 2,5 mg/kg) volatilized from the soil during 42 days (the trimethylselenonium ion is an important urinary metabolite of dietary Se) [11]. Atmosphere is a great reservoir of Se [12], composed from anthropogenic (62.5 %) and natural (37.5 %) sources. Vicinity of ocean can increase Se content in soil [13]. It is expected that the amount of Atlantic rainfall could effect on atmospheric emission of Se.

## Materials and Methods

Groundwater (gw) silicon (Si), calcium (Ca), magnesium (Mg) and potassium (K) values are provided by Geologic Survey of Finland [14]. Rural Centes (RC) '(04a). Finska Hushällningss.' and '(04b). Åland' are excluded, because of missing Se.H<sub>2</sub>O values and '(16).Ostrobothnia' because of small number of Si.gw samples and sulfurous soil [15]. Cropland hot water extractable Se values (Se.

H<sub>2</sub>O), ( $\mu\text{g}/\text{l}$ ) (N.B. weight per volume), in 1978-80 are from [16] (Soil fertility Service, Eurofins Viljavuuspalvelu Oy). (Method: dry and milled soil sample was extracted with boiled water at ratio 1:3. leachate was analyzed using CV-AAS equipment) [17]. Original data, (N=1340), were missing values concerning RC's (04a) and (04b). Exclusion of "(16).Ostrobothnia' caused ca 5 % (N 27) reduction, approximated by its cropland area of Finnish total value. Whole country mean of [Se.H<sub>2</sub>O] was 6  $\mu\text{g}/\text{l}$ . [Se.H<sub>2</sub>O] values by humus content were as follows: 0-3 %: 5  $\mu\text{g}/\text{l}$ , 3-12 %, 6  $\mu\text{g}/\text{l}$ , 12-20%: 7  $\mu\text{g}/\text{l}$ , by peat 4  $\mu\text{g}/\text{l}$  (remarkable is that values are per volume). Geographic coordinates of the Rural Centers are attained by web search: 'name of the visually selected central commune' and 'geographic coordinates' or "geographic coordinates dateandtime.info". In RC. (07) Renko has been after 1980 combined with Hämeenlinna, but data for both are accessible, why the old value was benefited. The order of Rural Centers is the same as by Eurofins Viljavuuspalvelu Oy. By order we must be careful with RC's (06), (07), (12) and (13) – it is not always the same!

Relative regional proportion of Atlantic rain, determined by change in 180/16O isotope ratio to respective Oceanic standard (VSMOW),  $\delta^{18}\text{O}$  (‰), has been estimated by combining the RC map (in [14]) and map in [18] Figure 1. Data for this study is in Table1.



**Figure 1:** Represents regional  $\delta^{18}\text{O}$  values by Rural Centers. Numbers of RC's are as given in [14], but RC.(04) is divided into two parts with different mineral element database (to clarify other articles).

**Table 1:** Names of Rural Centers after exclusion, names of central communes, groundwater Si, Ca, Mg, K, (Ca+Mg).

(Ca+Mg), Hot Water Extractable Se (Se.H2O), $\delta$ 18O, Latitude, Longitude and Altitude of Central Communes (As Given)												
	Central commune	Si.gw	Se.H2O	$\delta$ 18O	Latitude	Longit	Altitude	Ca	Mg	K	(Ca+Mg).gw	(Mg+K).gw
		mEq/l	$\mu$ g/l	‰	°N	°E	m			mEq/l		
01.Uusimaa	Järvenpää	1.14	9	-12.30	60.5	25.1	51	1.03	0.601	0.105	1.63	0.71
02.Nyland Svenska	Espoo	1.08	6	-12.20	60.2	24.7	16	0.89	0.379	0.090	1.27	0.47
03.Varsinais-Suomi	Aura	1.12	8	-11.80	60.7	22.6	51	1.05	0.601	0.159	1.65	0.76
05.Satakunta	Noormarkku	0.98	7	-12.30	61.6	21.9	44	0.82	0.288	0.184	1.11	0.47
06.Pirkanmaa	Ylöjärvi	0.93	6	-12.45	61.6	23.6	107	0.59	0.263	0.171	0.85	0.43
07.Kanta-Häme	Hämeenlinna	1.12	8	-12.30	60.9	24.3	107	0.89	0.543	0.115	1.43	0.66
08.Päijät-Häme	Asikkala	0.98	8	-12.60	61.2	25.5	74	0.88	0.362	0.113	1.24	0.47
09.Kymenlaakso	Anjalankoski	1.14	6	-12.70	60.8	26.8	18	0.79	0.321	0.079	1.11	0.40
10.South Karelia	Joutseno	0.93	6	-13.10	61.1	28.5	80	0.76	0.296	0.118	1.05	0.41
11."Mikkelin läänin" RC	Juva	0.91	6	-13.30	61.9	27.9	98	0.84	0.247	0.143	1.09	0.39
12.North Savo	Maaninka	0.78	5	-13.40	63.2	27.3	88	0.65	0.206	0.102	0.85	0.31
13.North. Karelia	Kontiolahti	0.81	6	-13.70	62.8	29.9	138	0.55	0.181	0.138	0.73	0.32
14.Central Finland RC	Saarjärvi	0.90	5	-13.10	62.7	25.3	140	0.49	0.156	0.090	0.65	0.25
15.South Ostrobothnia	Nurmo	1.07	6	-12.80	62.8	22.9	47	0.68	0.247	0.212	0.93	0.46
17.Central Ostrobothnia	Toholampi	0.90	5	-13.20	63.8	24.3	94	0.56	0.189	0.092	0.75	0.28
18.North Ostrobothnia	Ylikiiminki	0.85	5	-13.45	65.0	26.2	80	0.79	0.280	0.169	1.07	0.45
19.Kainuu	Ristijärvi	0.65	4	-13.70	64.5	28.2	160	0.67	0.214	0.087	0.88	0.30
20.Lapland	Rovaniemi	0.70	4	-14.40	66.5	25.7	92	0.56	0.230	0.100	0.79	0.33

## Results

### Comments on Table 2

All associations of [Si] and [ $\delta$ 18O] were significant. Long and Alt explained [Se.H2O] weakly: by 14.5 % (ns) and respectively by 17.3 % ( $p < 0.05$ ). Combined regression by [Lat;Long;Alt] explained

better [Si] (by 84 %), Figure 2, than [Se.H2O] (by 65.2 %) Figure 3. [ $\delta$ 18O] behaved rather similarly. [Ca+Mg] and [Mg+K] explained stronger [Se.H2O], (by 68.9 %, Figure 4, and 72.2 %, Figure 6, respectively,  $p < 0.001$ ) than [Si], (by 52.1 %, Figure 5, and 53.9 %, Figure 7, respectively,  $p < 0.01$ ). [Se.H2O] was explained 62.7 % by [ $\delta$ 18O] and 61.9 % by [Si] Figure 8, respectively ( $p < 0.001$ ).

**Table 2:** Regressions of groundwater Si, [Se.H2O] and  $\delta$ 18O by Latitude, Longitude, Altitude and groundwater.

(Ca, Mg) And (Mg+K), R Squares and Significance Levels (Ns: Non-Significant; *: $P < 0.05$ ; **: $P < 0.01$ , *** $P < 0.001$ )			
	Si	Se.H <sub>2</sub> O	$\delta$ 18O
Lat	67.5*** (-)	61.0*** (-)	74.2*** (-)
Long	27.6* (-)	14.5 ns (-)	41.9** (-)
Alt	49.3** (-)	17.8 (*) (-)	32.3* (-)
[Lat;Long]	79.2 *** (-68;-32)	65.2*** (-78;-22)	95.2***(-61;-39)
[Lat;Long;Alt]	84.0*** (-54;-22;-24)	65.4*** (-72;-22;+6)	95.2***(-60;-38;+2)
(Ca+Mg)	52.1** (+)	68.9*** (+)	53.0** (+)
(Mg+K)	53.9** (+)	72.8*** (+)	56.4*** (+)
$\delta$ 18O	75.5*** (+)	62.7*** (+)	100
Si	100	61.9*** (+)	

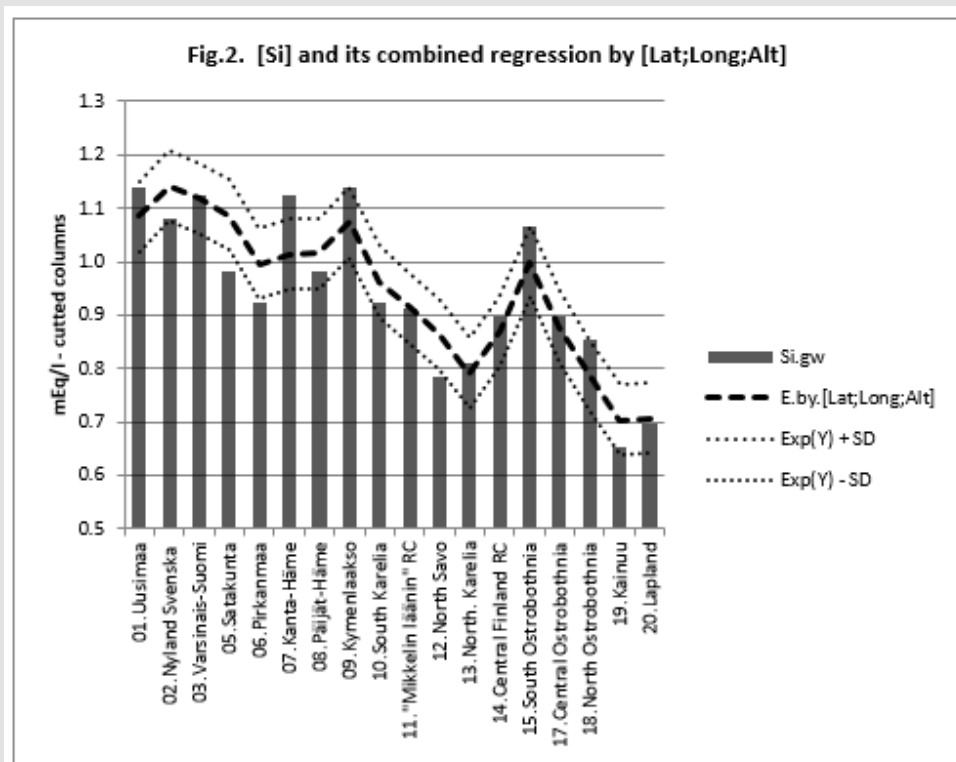


Figure 2: Represents groundwater Si and its regression by geographic factors [Lat;Long;Alt].

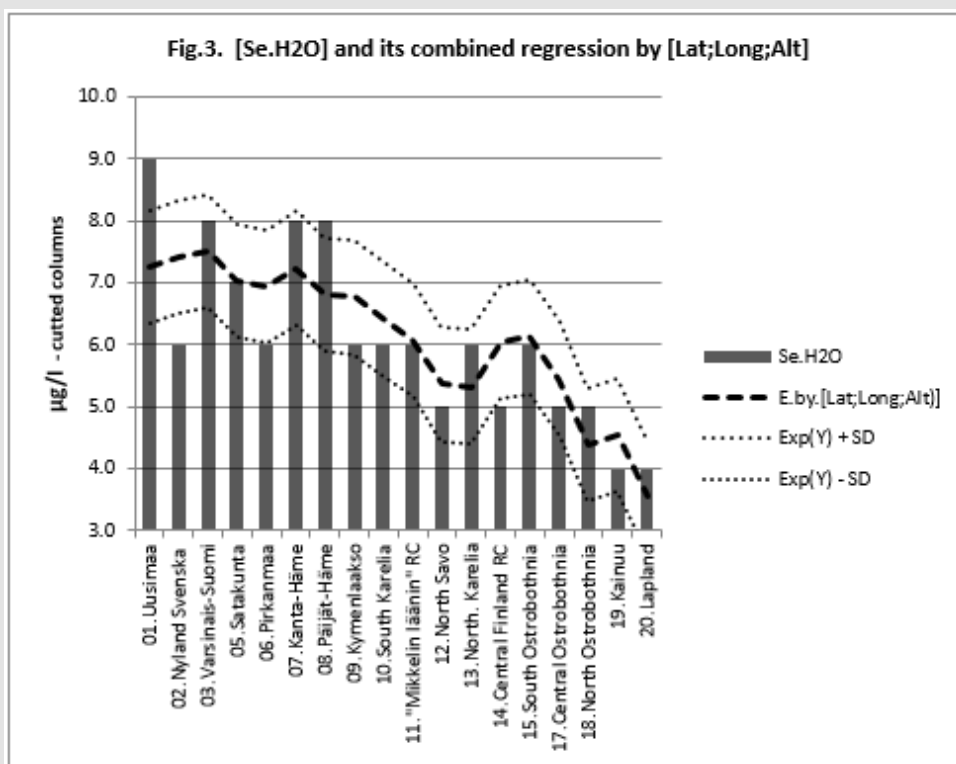


Figure 3: Represents cropland [Se.H2O] and its regression by geographic factors [Lat;Long;Alt].

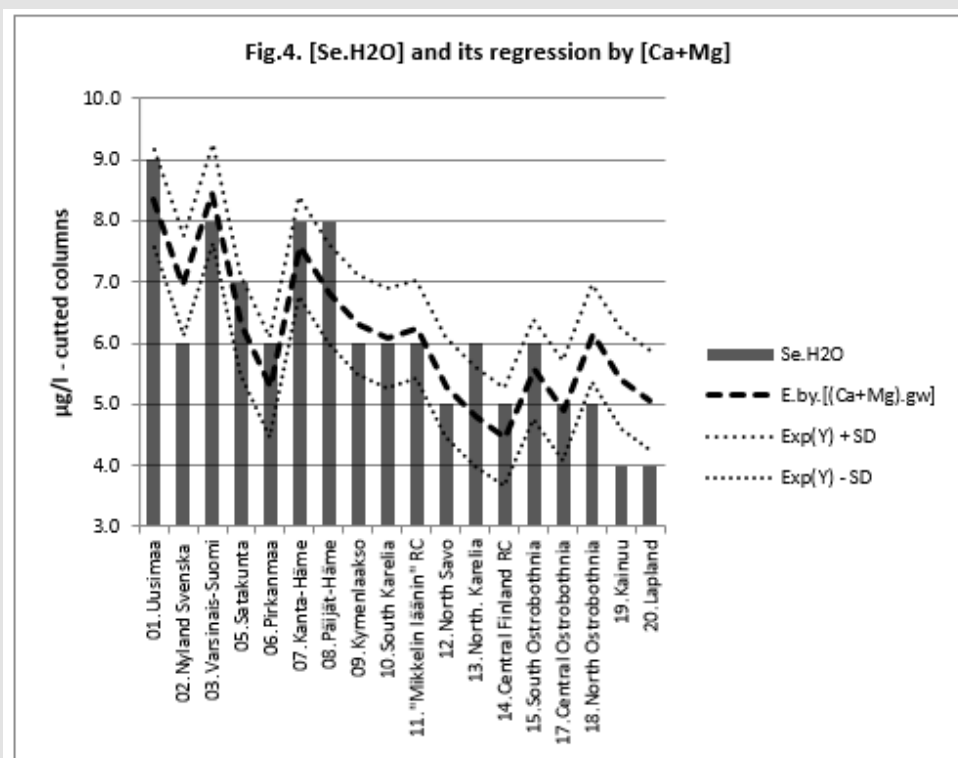


Figure 4: Represents cropland [Se.H2O] and its regression by groundwater (Ca+Mg).

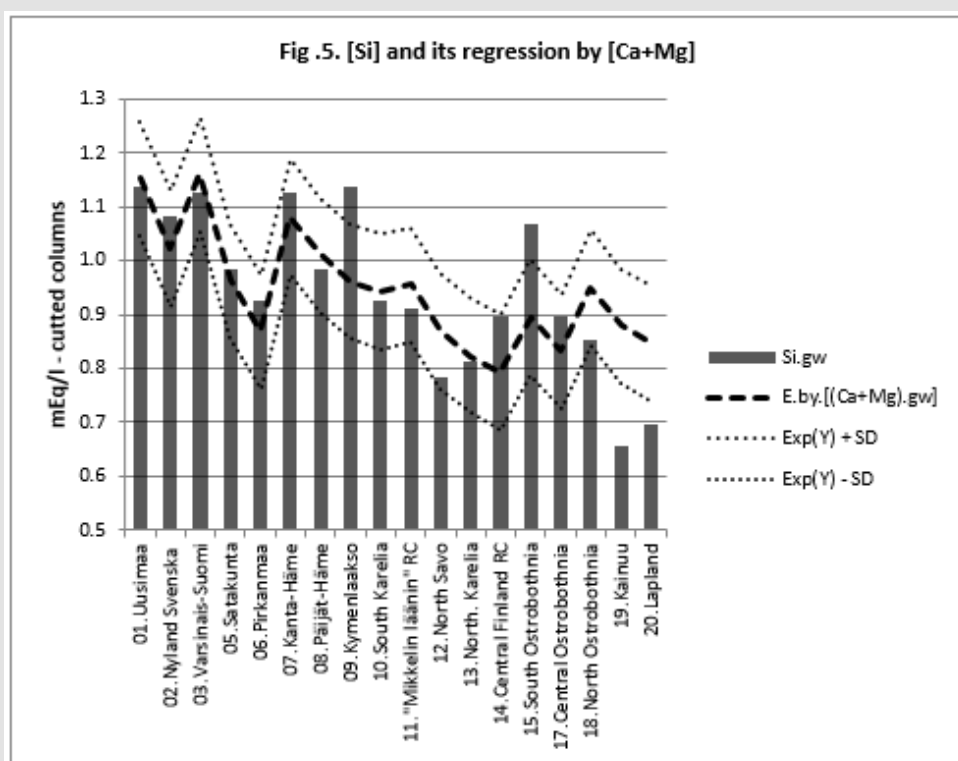


Figure 5: Represents groundwater Si and its regression by groundwater (Mg).

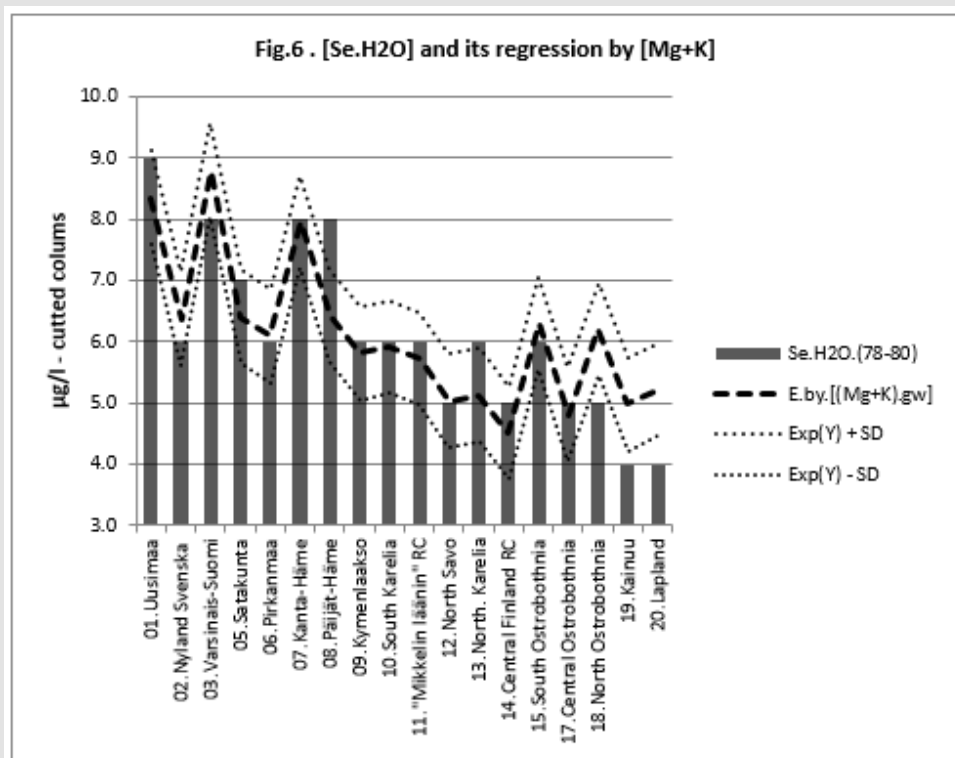


Figure 6: Represents cropland [Se.H2O] and its regression by groundwater (Mg+K).

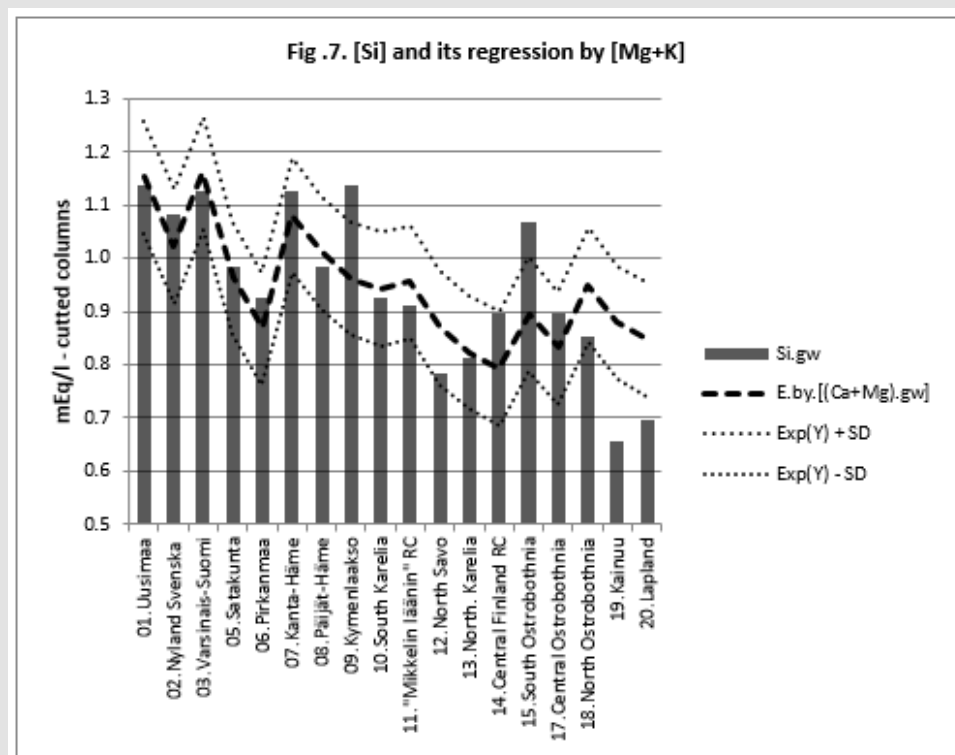


Figure 7: Represents groundwater Si and its regression by groundwater (Mg+K).

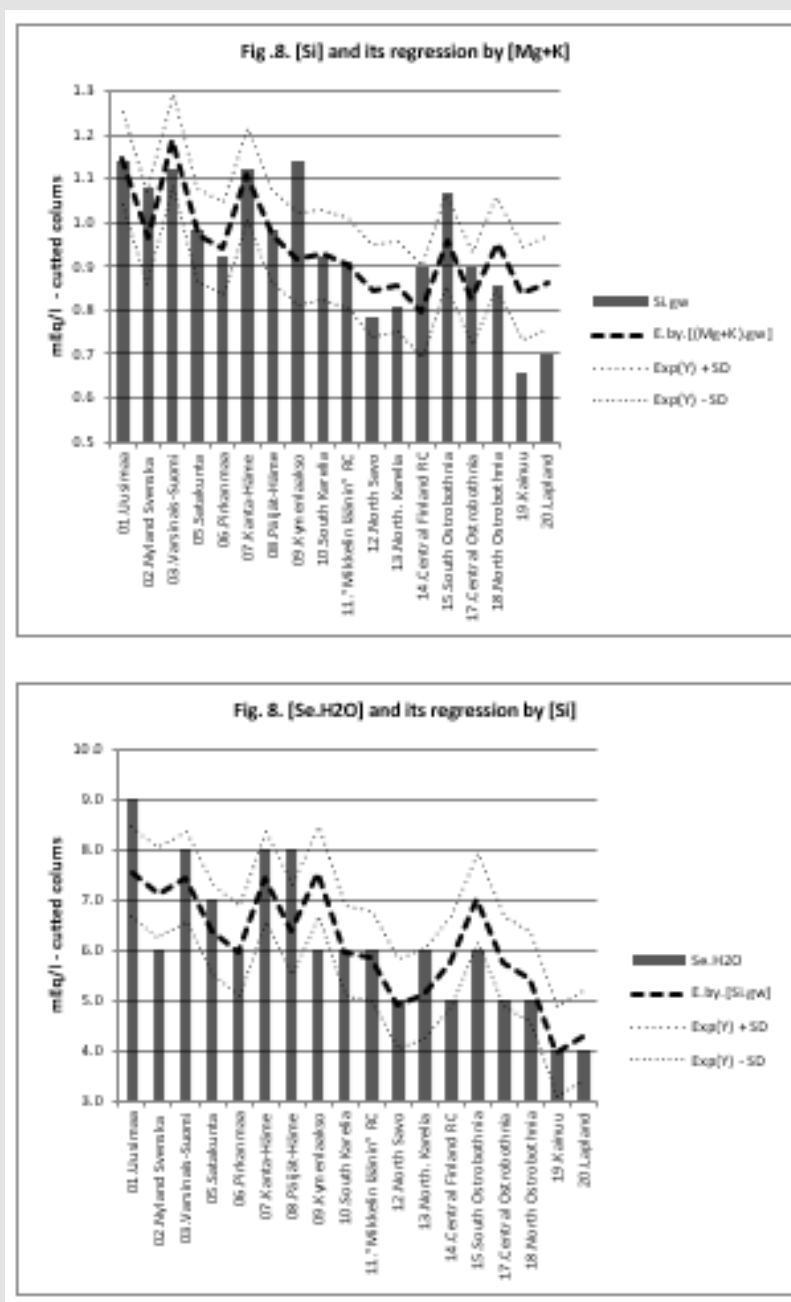


Figure 8: Represents [Se.H2O] and its regression by groundwater Si.

### Discussion

Remarkable are the high/remarkable associations with geographic factors, especially by [Alt], which has been determined by one point of central commune, which is only a part of each Rural Center. Weathering of rocks in mountains produces sediments that are transported downstream by erosion to create fertile soil in lower parts of the landscape [19]. [Lat], [Long] and [Alt] can be seen as erosion factors: Latitude is associated with temperature

(Finnish range of latitude is from 60 to 70 °N). Longitude in general indicates distance to Baltic Sea and soil age [20]. [Alt] means more loss than receiving of minerals via erosion. Combined regression by [Lat;Long;Alt] explained [Si] 84.0 % (p < 0.001), [Se.H2O] 65.4 (p < 0.001) and [ $\delta^{18}O$ ] 95.2 % (p < 0.001). Single associations by these “erosion/dilution factors” were negative, but in combined regressions coefficients of [Alt] were positive by [Se.H2] and by [ $\delta^{18}O$ ], which could suggest, that the losses by erosion could have been partially compensated.

[Se.H<sub>2</sub>O] was strongly explained by [Mg+K] (72.8 %,  $p < 0.001$ ), more strongly than [Si] (53.9 %,  $p < 0.01$ ). [Se.H<sub>2</sub>O] association can be understood by its association with biotite and clay [5,6]. It even suggests that [Si] has other important sources besides biotite, too. [Si] explained [Se.H<sub>2</sub>O] by 61.9 % ( $p < 0.001$ ). This can be understood by [Se.H<sub>2</sub>O] -humus [16] and humus-[Si] [21] associations. Associations of [Se.H<sub>2</sub>O] are surprisingly high, range of values is from 4 to 9 ( $\mu\text{g/l}$ ), and values are integers. [Se.H<sub>2</sub>O] sample collection, with regional analyses, [16] from period before Se fertilization is worthy of attention. When the results (by Rural Centers) were composed to provincial [Se.H<sub>2</sub>O] values (not presented here), they explained stronger timothy Se in [2]. than “soluble” (AAAc-EDTA extractable) Se in [2]. Remarkable is the increasing trend in the hot water extractable [Se.H<sub>2</sub>O] values: 6  $\mu\text{g/l}$  [16] in 1978-80, 7.3  $\mu\text{g/l}$  in 1982-84 [22] before Se fertilization, 7.8  $\mu\text{g/l}$  in 1985-89 (during Se supplementation) [22], (based on data of Soil fertility Service as [16], with sampling time usually autumn [23]), 10  $\mu\text{g/l}$  in 1998 [24] (in early summer, when “timothy had formed a full spike”, ca one month after Se supplementation) and 9  $\mu\text{g/l}$  in 2006 (estimated mean by author from [1], where Se content in sand was 7  $\mu\text{g/kg}$  and in clay 13  $\mu\text{g/kg}$  (by rough volume weight estimate 1 by [4], ca 10-12 months after Se fertilization). Data in [16], as well as in [22] are results of commercial analyses.

There are several studies on Se content in soil and plants. But the number of samples has been generally small. “Soluble Se” can be understood at least in three ways:

- a) water-extractable,
- b) AAAc-EDTA extractable and
- c) acid ammonium oxalate extractable

Additionally, water extraction can be performed by shaking or boiling. Sampling time-point could affect on results during Se fertilization: e.g. 1 or 12 months after fertilization [24,1]. Label “soluble Se” is non-precise and misleading, e.g., in [4]. N of samples in [16] (1340) is higher than in [2] (250), which obviously made the values less sensitive on environmental factors, why it associated better with timothy Se [2] (can be calculated). (Environmental factors are not assessed in this article, because specific data on environmental factors concerning [Se.H<sub>2</sub>O] is difficult to find). Availability of Se from fertilizers to plants disappears rapidly after fertilization [25], why [Se.H<sub>2</sub>O] increase between 1980-2006 suggests on change in active Se reservoir [16,1]. High [Se.H<sub>2</sub>O] variation in the 1980's [22] could be explained by sampling and moderately low Se in early summer in 1998 additionally by environmental factors. Possibly rapid turnover of [Se.H<sub>2</sub>O] could partially explain why increase in [Se.H<sub>2</sub>O] is lower than in plant Se. Anyhow 50 % increase in “plant-available” [Se.H<sub>2</sub>O] [16,24] could not predict the possible 30-500-fold increase in plant Se [25] caused by fertilizers.

In Finland there are no satisfactory studies on regional airborne Se depositions. In 1990 Finnish total “anthropogenic” Se fallout from precipitation was approximately 18.4 t/a (0.54 g/ha) [26]. Se content in rain was 118 ng/l, and in snow 63.1 ng/l, suggesting on moderate inaccuracy, because consumption of coal and oil was obviously higher during snowing than raining [26]. Support on (some part of) Atlantic Se fallout from precipitation gives Danish rain water (250 ng Se/l) in 1971 [26]. Se association with sulfur is known [26]. So higher sulfur emissions in 1978-80 to 1991 [27] together with inaccuracy in Se determination [26] could suggest on availability of airborne upto Se 1 g/ha/a in 1978-80, cf. 12 g/ha/a via fertilizers in 1992-2004 [4]. The separate role of airborne Atlantic Se, possibly upto 1/3 of Se fallout [12], could not be determined. Anyhow all atmospheric Se via (common) southwest wind could have had compensated the Se losses – better than by Si - of the hills, which are impoverishing by erosion Table 2. Low content of molybdate-reactive silicon (1 %) in biotite extracts by oxalate [7] can be dependent on aluminium-silicon interactions [29] in acid solution (pH 0.65).

## Conclusions

During the time before Se supplementation “plant available Se”, [Se.H<sub>2</sub>O], obviously worked well. Humus is the home of soil biota, amount of “plant available Se” can be increased 10-15-fold by mycorrhizae [28], which explains the high association between humus and [Se.H<sub>2</sub>O]. Conclusions: [Si.gw] was more strongly than [Se.H<sub>2</sub>O] associated with geographic factors, but [Se.H<sub>2</sub>O] with clay-indicator [Mg+K], as well as with [Ca+Mg]. Positive coefficient of [Se.H<sub>2</sub>O] with [Alt] in combined regression by [Lat;Long;Alt] suggests “hint-like” that the altitude-erosion of Se could have been compensated by atmospheric Se. High association between [Si] and [Se.H<sub>2</sub>O] can be explained by their associations with humus. Se fertilization seemed to have influence on [Se.H<sub>2</sub>O] and its ability to predict plant Se.

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