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Influences of liming on the soil-plant transfer of Ra-226 from acid soils in field experiments

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Abstract

The effects of liming of the plough layer in the early sixties on the contents of exchangeable Ca and Ra-226 in soil and on the contents of Ca and Ra in the crops in the early eighties has been investigated. It was found that liming, while increasing the amounts of Ca, reduced the amounts of Ra and the ratio Ra/Ca exchangeable in soil. Liming influenced the plant upake of Ra more for the vegetative than for the generative parts of the grain crops. However, the reduction of the Ra/Ca-ratio in the former was not as effective as in the soil. In the grain it was uncertain. The difference can, however, be explained by the fact that the minerals in straw and grain are more or less taken from different layers of/soil profile. The crop is more dependant on the plough layer during the early development, than later when grain is developed.

Introduction

The soil development in humid regions produces with time through natural processes leached soil profiles, with different horizons more or less acidified and depleted with life supporting elements. The agricultural soils are generally more resistant to acidification than the forest soils now in danger. However, a considerable leaching of the former is going on, (1, 2) and natural soil and climatic conditions as well as artificial means (like agricultural production and harvesting, and fertilization in addition to acidified precipitation) has contributed to the present diverse state of our soils with regard to their content of bases and to their pH-levels (2, 3). The soil calcium, in some cases present as carbonate but in most cases sorbed to the soil exchange complex, determines the properties of the soil and its agricultural value. It also influences the transfer of heavy metals to the crop as the ratio M/Ca in soil influences the ratio in the crop.

Now, during the leaching process it can be expected from chemical reasons that relatively more calcium than heavy metals like Cd and Ra is leached out of the soil profile and the plant root horizon. As a consequence the ratio of the exchangeable amounts of Ra and Ca, Ra/Ca, in soil will increase with time and cause an increased contamination of Ca in plants with Ra. The general doubling time can only be speculated on. One thing is certain, however, the rate of change is increasing due to the increasing intensity in some of the factors governing the acidification process.

To monitore this process and its effect is not possible, as it should require more than a decade of analythical observations. However, if the acidification process goes on the effects are unavoidable. The only remedy known is liming of agricultural land, and below is given some preliminary data from 5 field experiments started during the early sixties. Samples of soils and crops have been obtained in a cooperation project with the Department of Soil Sciences at the Swedish University of Agricultural Sciences, Uppsala(cf. 4).

Materials and methods

Detailed data on the soils of the experimental sites and on the outcome of the liming treatments can be obtained from (4). Some of the soil data are given in Table 1. They show that the common original characteristics are the low pH-levels and the very low to low Ca-saturation level of the cation adsorption complex. Otherwise the soils are different. They represent sandy soils to loams with moderate clay contents.

The contents of uranium, and of radium in the unlimed treatment are given in Table 2. These contents may be regarded as representative for a low to moderate level, in the Swedish soils as compared to those in Appendix A.

The methods for analysis of uranium and ^{226}Ra are described elsewhere (5). Determination of "Rn_p" is, however, recently developed. "Rn_p" or "the potential Rn-exhalation capacity" is a concept equivalent to a fraction of ^{226}Ra in soil which exhales ^{222}Rn possible to determine by comparison to the emanation from a standard ^{226}Ra-source. The procedure is the following: 10-20 g dry soil is added to a 100 ml glass bottle with 0.01 M NaOH. The bottle is shaken and left over night for dispersal of the soil aggregates. Then 6 ml n-Hexan is added, the bottle is filled with aq dest., allowing for a 0.5 ml air bubble, and sealed and kept for a period of 30-40 days. After this period the bottle is shaken vigorously for one hour to release the Rn produced from the solid phase and to allow for equilibrium to be attained between the radon in the water and in the organic phase. After that an aliquote of the hexan phase is taken and mixed with 10 ml of a liquid scintillator, left for cooling and measured after 3-4 hours when the radon and the daughters are in equilibrium.

Results and discussion

The experimental soils were sampled 15-16 years after start and analyzed. Data for the content of exchangeable amounts of Ca and Ra, and Rn_p are given for the three liming treatments on the five soils in Table 3. As was to be expected the liming had increased the content of exchangeable Ca in the soil. However, another effect was that the amount of exchangeable Ra had decreased, and as a consequence of these changes the ratio (Ra/Ca) exch. was greatly reduced. The Rn_p-values was not influenced by any treatment and no loss of radium from the soil was indicated. The Rn_p represents a larger fraction of the soil radium comparable to that extractable with hot 7 M HNO_3 (cf. App. A).

The effects of liming on the mineral contents of the crops in 1982 are given in Table 4. In common for Expts 1, 2 and 4 is the fact that liming has increased the content of Ca and reduced the content of Ra and the ratio Ra/Ca in the vegetative parts of the crops. In Expt. 3 the effect is uncertain and in Expt. 5 the straw was not available for analysis.

The liming effects on the generative parts of the crops, the grain is smaller and less certain, at least in 1982. Such an observation for the Ca-content is natural. However, the reduced Ra/Ca-ratios in the limed soil treatments of the Expts should have had effects also on the Ra/Ca-ratios in the grain. One explanation might be that radium is discriminated compared to calcium during the transport upwards through the stem. However, higher Ra/Ca-ratios in grain than in straw has been observed (6) and can be found also in App. D. This aims at another explanation. The minerals in straw and in grain may come from different sources or levels in the soil profile as 20-80 % of plant calcium may be taken from the subsoil (7). On the experimental sites the whole soil profiles available to plant roots are acidified, and the liming has improved only the plough layer. The subsoil below all treated top layers as well as below the control must be largely the same and not much influenced by the liming. As the plants depending on soil and on weather conditions more or less feed in the subsoil layer the liming effect on the Ra/Ca-ratio in plants is conditional. It is higher in the vegetative parts, built up during the early part of the season when the plant feeds more in the top layer. The effect is lower in the generative parts built up during the latter part of the season when the root systems have been developed to extract water and nutrients from horizon below the drier upper parts of the soil profile.

This implies that the liming intentions is not fulfilled in every respect after only a few decades. It improves the yield in the short run, but not always the quality. So is i.e. the uptake of Cd (8) from acidified soil profiles not reduced nor that of radium in grain for human consumption.

The crops are different in all the five experiments. This makes it difficult to rank the soils unless the crops are ranked on beforehand. A comparison to App. B, C and D shows that the Ra-content and the Ra/Ca-ratio in grain decrease in the order wheat > barley, rye > oats. From this follows that the radium transfer from the soils to the crops seems to decrease in the order 2, 5 > 3, 4 > 1. This ranking is confirmed when compared to the OR-values calculated in Table 5. However, only those calculated for the control treatment I should be considered as the liming after 15 years has improved only the top soil layer. Then the consistency of the OR-values seems acceptable although the transfer of radium from Soil 2 to the crop is relatively high and the discrimination of radium by oats is outstanding. The total range in OR for vegetative plant parts is found to be 2×10^{-2} -- 9×10^{-2} and that for grain 1×10^{-2} --6×10^{-2}.

Calculation of TF_{sp} (Table 5) has a meaning for Ra_{tot} and Rn_p only if those fractions are interrelated to Ra_{exch} or the plant available Ra-fraction. Due to influence of calcium on the plant uptake of radium they are less consistent than OR-values.

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Table 1. Soil characteristics at the start of the field liming experiments in 1964 (cf. 4).

Soils		Soil type	pH_{aq}	CEC, me per 100 g soil	Ca-saturation %	K_{HCl}, mg per 100 g soil
Amalietorp	1	Fine sand	5.4	17	55	46
Tönnersa	2	"-	5.5	6	50	82
Ulfstorp	3	Loam	5.7	20	60	180
Eckerud	4	" , gy	4.7	24	10	144
Säby	5	"	5.8	23	65	260

Table 2. Total content of uranium and radium acc. to analysis by fusion of 1 g soil with sodium carbonate and the "potential Rn-exhalation capacity" $^{222}Rn_p$ [a)].

Soil No	mBq per g dry soil ^{238}U	^{234}U	^{226}Ra	$^{222}Rn_p$	Rn_p/Ra
1	43	51	46	17 ± 1.7	0.4
2	17	18	24	5 ± 0.6	0.2
3	60	63	53	37 ± 2.3	0.7
4	44	40	44	27 ± 2.2	0.6
5	59	71	55	39 ± 1.1	0.7

[a)] determined with 10 g dry soil

Table 3. Ca and ^{226}Ra, exchangeable with 1 M NH_4Cl. 1:10, and $^{222}Rn_p$ [a)], in the liming treatments[b)] of the field experimental soils. Averages for 3 phosphate fert. levels and for 2 levels of crop residues ploughed in, 0 and 1.

Soil	Treat-	Ca		^{226}Ra		Ra/Ca		$^{222}Rn_p$	
No	ment	mg/100 g	CV %	Bq/kg	CV %	Bq/g	CV %	Bq/kg	CV %
1	I	119	26	4.1	10	3.6	25	16	14
	II	169	2	4.7	21	2.8	18	18	7
	III	228	3	3.6	18	1.6	20	17	11
2	I	21	10	2.4	7	11.8	16	5	5
	II	41	4	2.1	3	5.2	2	5	20
	III	56	35	1.4	8	2.9	48	5	11
3	I	177	5	20.4	6	11.5	11	37	10
	II	215	10	16.2	24	7.5	22	38	2
	III	356	5	9.7	22	2.8	26	36	1
4	I	72	7	15.0	11	21.0	7	25	9
	II	281	9	13.4	3	4.8	10	26	4
	III	374	1	12.2	3	3.3	2	29	6
5	I	218	12	23.1	5	10.7	15	38	3
	II	254	4	21.7	4	8.6	6	40	3
	III	376	1	16.1	15	4.3	16	39	2

a) $^{222}Rn_p$ = "potential Rn-exhalation capacity"

b) I Unlimed

II Liming up to 70 % - saturation level in 1964

III " " " 100 % - " " " "

Table 4. Content of radium and calcium in crops from the liming experiments in 1982.

Expt. Crop	Treatment No.		Content per kg dry matter Ra, Bq Grain	Ra, Bq Straw	Ca, g Grain	Ca, g Straw	Ratio, Ra/Ca Bq/g Grain	Ratio, Ra/Ca Bq/g Straw
Amalietorp 1	I			1.1		9.3		0.15
(Hay)		CV %		26		40		62
	II			1.0		11.5		0.08
		CV %		25		20		26
	III			0.7		12.1		0.06
		CV %		28		23		42
Tönnerstad 2	I		0.5	2.1	0.7	2.0	0.7	1.1
(Winter rye)		CV %	27	26	6	8	26	25
	II		0.5	1.7	0.9	2.7	0.5	0.6
		CV %	17	20	20	5	15	16
	III		0.4	1.4	0.8	3.2	0.4	0.5
		CV %	24	24	12	7	24	32
Ulfstorp 3	I		0.3	1.4	0.8	2.1	0.4	0.6
(Winter wheat)		CV %	30	32	18	6	32	36
	II		0.3	1.5	0.8	2.3	0.4	0.7
		CV %	31	21	7	6	33	23
	III		0.3	1.2	0.8	2.2	0.4	0.6
		CV %	31	41	5	12	30	36
Eckerud 4	I		0.2	1.1	1.2	3.9	0.1	0.3
(Oats)		CV %	31	14	15	14	39	5
	II		0.1	1.4	1.3	6.2	0.1	0.2
		CV %	39	21	8	11	39	35
	III		0.1	0.7	1.4	6.8	0.1	0.1
		CV %	38	37	10	11	39	35
Säby 5	I		0.5		0.9		0.6	
(Barley)		CV %	13		7		17	
	II		0.5		0.9		0.5	
		CV %	27		9		28	
	III		0.4		0.9		0.5	
		CV %	28		4		25	

Table 5. Calculated transfer factors based on dry matter weights and different fractions of ^{226}Ra in soil, $TF_{sp} \times 10^{-2}$ and the observed ratios $(Ra_p/Ca_p)/(Ra_s/Ca_s)_{exch}$ OR $\times 10^{-2}$.

			Grain $\times 10^{-2}$				Straw $\times 10^{-2}$			
			TF_{sp}			OR-	TF_{sp}			OR-
Soil No	Treatment	Crop	Ra_{tot}	Rn_p	Ra_{exch}	value	Ra_{tot}	Rn_p	Ra_{exch}	value
1	I	Hay	-	-	-	-	2.4	6.5	27	4
	II		-	-	-	-	2.2	5.9	21	3
	III		-	-	-	-	1.5	4.1	19	4
2	I	W.rye	2.1	10	21	6	8.8	42	88	9
	II		2.1	10	24	10	7.1	34	81	12
	III		1.7	8	29	14	5.8	28	100	17
3	I	W.wheat	0.6	0.8	1.5	4	2.6	3.8	7	5
	II		0.6	0.8	1.9	5	2.8	4.1	9	9
	III		0.6	0.8	3.1	14	2.3	3.2	12	21
4	I	Oats	0.5	0.7	1.3	1	2.5	4.1	7	2
	II		0.2	0.4	0.7	2	3.2	5.2	10	4
	III		0.2	0.4	0.8	3	1.6	2.6	6	3
5	I	Barley	0.9	1.3	2.2	6	-	-	-	-
	II		0.9	1.3	2.3	6	-	-	-	-
	III		0.7	1.0	2.5	12	-	-	-	-

App. A. Distribution of 60 cultivated soils (sampled in 1976 in the County of Örebro) on 3 classes with regard to the size of different fractions of ^{226}Ra (1976).

Fraction of Radium	Low Bq/kg	Low %	Medium Bq/kg	Medium %	High Bq/kg	High %
Ra_{tot}	<50	≈23	50-100	≈61	>100	≈16
Ra_{HNO_3}	<50	≈44	50-100	≈41	>100	≈14
Rn_p	<50	≈53	50-100	≈38	>100	≈9
Ra_{exch}	<10	≈32	10-30	≈61	>30	≈7
Ratios						
$(Ra/Ca)_{exch}$ Bq/g	<5	≈23	5-20	≈67	>20	≈10
Rn_p/Ra_{tot}	<0,5	≈13	0,5-0,8	≈70	>0,8	≈17

App. B, 1-2. Distribution of 100 field grain crops in Sweden on 5 classes with regard to the content of ^{226}Ra per kg dry matter and per g Ca. Sampling survey in 1978.

1. Class limits, Bq per kg dry matter		Wheat	Rye	Barley	Oats	Oilseed
1	<=0.1	2	0	10	11	2
2	>0.1-<=0.2	3	5	27	6	2
3	>0.2-<=0.3	3	0	7	1	4
4	>0.3-<=0.4	3	0	3	0	1
5	>0.4	3	0	3	0	4

(Number of each crop)

2. Class limits, Bq per g Ca		Wheat	Rye	Barley	Oats	Oilseed
1	<=0.1	0	0	7	9	7
2	>0.1-1=0.2	1	0	10	6	4
3	>0.2-<=0.3	2	2	17	3	1
4	>0.3-<=0.4	1	2	5	0	1
5	>0.4	10	1	11	0	0

(Number of each crop)

App. C, 1-2. Distribution of 72 field hay crops in Sweden on 5 classes with regard to the content of ^{226}Ra per kg dry matter and per g Ca. Sampling survey in 1978.

1. Class limits, Bq per kg dry matter		%	2. Class limits for Ra/Ca, Bq per g Ca		%
1	<=0.3	6	1	<=0.1	19
2	>0.3-<=0.6	36	2	>0.1-<=0.2	22
3	>0.6-<=0.9	25	3	>0.2-<=0.4	27
4	>0.9-<=1.2	15	4	>0.4-<=0.6	13
5	>1.2	15	5	>0.6	19
Total		100			100

App. D. Comparison between crops, with regard to the ratio Ra/Ca, Bq/g. Samples taken 1965-68 from the same field and in the same year in crop rotation experiments.

Farm and county	W.wheat		S.wheat		Barley		Oats		Hay crops	
	Grain	Straw	Grain	Straw	Grain	Straw	Grain	Straw	I	II
Ulfhäll D	1.00	1.60	0.45		1.17	1.10	0.18		0.32	0.14
Ultuna C	0.82	0.72	0.71	0.28	0.42	0.23	0.13	0.22	0.09	0.09
Borgeby M	0.34	0.06	0.35	0.05	0.28	0.06				
Lönhult M	0.77	0.45	0.46	0.40	0.33	0.07				
Flahult F	0.37[a]		0.34[b]	0.28[b]	0.80	0.20	0.34	0.24	0.10	
Ugerup L	0.79[a]	0.09[a]			1.17	0.18			0.09	0.13

[a] = Winter rye   [b] = Summer rye