

## **Calcium Budgets for Catchments as Interpreted by Strontium Isotopes**

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Strontium isotopes in precipitation, soil and runoff water can be used to establish a ratio of wet plus dry deposited Sr to Sr released by weathering. This ratio is especially enhanced in areas with old acid Proterozoic rocks (0.6-2.5 Ga) and Archean rocks (>2.5 Ga). Since Sr and Ca behave in an analogous way in the coniferous forest ecosystem the results for Sr can be used for the determination of Ca. If the deposition of calcium can be calculated reasonably accurately the weathering rate can also be estimated. Five catchments have been investigated using this approach. Three of them seem to be close to a steady state, wherein the losses and gains of calcium to the system are equal. In the two southernmost catchments there seems to be an ongoing loss of exchangeable calcium. The loss by runoff occurs with sulphate being the dominant anion. Weathering rates of 1.5 to 4.8 kg Ca/ha year have been estimated.

### **Introduction**

Some of the major elements in soils and waters are stored in considerable amounts in the soil in exchangeable form. If an ecosystem is subject to a disturbance these stores can act as buffers. For instance, buffering to acidification by atmospheric acid deposition can be neutralized by exchangeable cations until the stores are depleted (Paces 1986). To obtain a steady state condition the mobilization of base cations by weathering must match the acid input, at least over a forest generation. Calcium is the base cation which occurs in the largest amounts on exchange sites. Thus it appears important to distinguish between neutralization by exchangeable calcium and neutralization by weathering derived calcium.

Strontium and calcium exhibit a very similar behaviour in a coniferous forest ecosystem (Popovic and Nommik 1967; Åberg and Jacks 1989). Of the three major stable Sr isotopes,  $^{86}\text{Sr}$ - $^{87}\text{Sr}$ ,  $^{87}\text{Sr}$  may get an additional input from the radiogenic decay of  $^{87}\text{Rb}$ . The ratio  $^{87}\text{Sr}/^{86}\text{Sr}$  is thus dependant on the environment, *i.e.* how much Rb has been available and for how long a time? By determining the makeup of the Sr ratio different Sr sources can be characterized.

In seawater the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio is about 0.709 at present. Strontium has a residence time in the sea of about  $4 \times 10^6$  years, which when compared with the mixing rate of about  $10^3$  years results in an isotopic ratio for seawater that is homogeneous at any given time. In precipitation over Sweden the ratio is slightly higher, or about 0.715 (Åberg and Jacks 1989). In Swedish Proterozoic rocks the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio is elevated to a varying degree depending on age and type of rock from which the Sr is derived. The decay of  $^{87}\text{Rb}$  is quite slow (half-life *c.*  $5 \times 10^{10}$  years) a fact which favours the use of the strontium ratio in older material. Consequently, it is possible to distinguish strontium derived by atmospheric deposition from that mobilized by weathering in areas with Proterozoic ( $2.5 - 0.6 \times 10^9$  years) and Archean ( $> 2.5 \times 10^9$  years) rocks and soils derived from these rocks.

The ratio  $^{87}\text{Sr}/^{86}\text{Sr}$  in runoff water reflects the long term input ratio to an ecosystem of atmospherically derived strontium relative to strontium mobilized by weathering. As calcium and strontium behave in an analogous way the same can be said for calcium. If the calcium deposition can be calculated with a fair accuracy then the weathering rate can be estimated. If uptake by trees and export by runoff is known a complete budget can be established.

## Materials, Methods and Sites of Investigation

The methodology of the sample collection and analytical work has been presented in previous papers (Åberg and Jacks 1989; Wickman and Åberg 1987). The soil samples were analyzed on an Avco 901 mass spectrometer at the Laboratory for Isotope Geology, Stockholm, and the water samples on a Micromass 54 E mass spectrometer at Scottish Universities Research and Reactor Centre (SURRC), East Kilbride. The analytical error is less than 0.05% and 0.01%, respectively. Our internal standard analyzed in East Kilbride (Stockholm tap water of 1983-10-27) gave a Sr value of 0.72696 to be compared with a previous Stockholm mean value of  $0.72705 \pm 0.00007$  (triplicate decompositions, Wickman and Åberg 1987).

In this paper results will be presented from the five catchments (Fig. 1) which are described in more detail by Tamm (1925), Rosén (1982), Grip (1982) and Nihlgård (1985). Some important characteristics are summarized in Table 1. The analytical values for the soil and water samples are compiled in Tables 2 and 3, respectively.

The Svartberget and Kullarna (Övre + Nedre) catchments (Fig. 1) are underlain by gneisses and gneissic granites, intermediate in composition. Plagioclases domi-

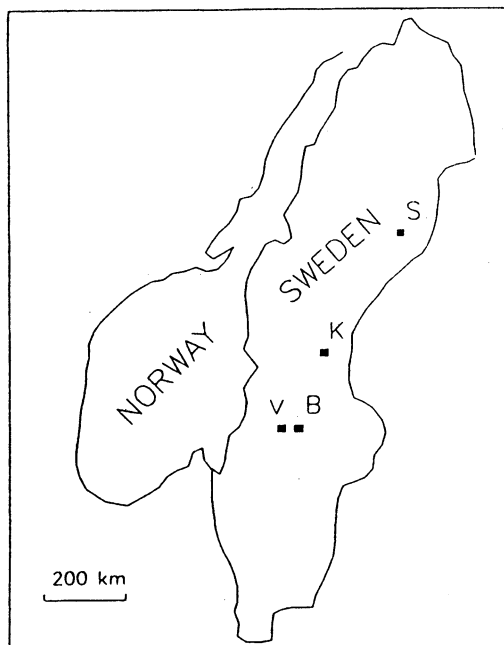


Fig. 1. Location of the Svartberget (S), Kullarna Övre and Nedre (K), Buskbäcken (B) and V Dybäcken (V) areas.

nate over potassium feldspars and there is about a 5-10 % dark mineral content in the soils derived from the rocks. In Buskbäcken and V Dybäcken (Fig. 1) acid granites form the basement. In the soils, potassium feldspars dominate largely over plagioclases (obligoclase) and there are only rather small amounts of dark minerals present. The dark minerals are generally hornblendes when observed in thin sections under the microscope.

For the budgets, records of varying length have been used, longest for the Kullarna (Övre + Nedre) catchment and shortest for the V Dybäcken catchment (14 and 2 years respectively). The depositions were calculated by using measured precipitation, sometimes corrected for elevation (7%/100 m difference) and multiplying by the concentration of calcium recorded in the atmospheric chemistry network (operated by the Meteorological Department, Stockholm University).

This procedure has been found to be adequate for sites between the stations in the network (Rodhe and Granat 1984). Dry deposition was calculated for the Buskbäcken catchment in a previous report (Åberg and Jacks 1989) by using aerosol measurements from a nearby site (Lannefors *et al.* 1983). A dry deposition rate of calcium representing 20 % of the wet deposition was obtained. The same percentage was used for V Dybäcken, while 15 % was used for Kullarna (Övre + Nedre) and 10% for Svartberget. In general it is found that dry deposition relative to wet deposition decreases northwards over Sweden (Monitor 1981). This maybe true also for calcium as the source areas are agricultural activities in southern

Table 1 – Location, size and type of the investigated catchments

Catchment	Lat.	Long.	Elevation m.a.s.l.	Area ha	Soil	Vegetation
Svartberget	64°15'	19°45'	240-310	53	till	spruce-fir
Kullarna Ö	62°00'	16°34'	310-440	50	till	spruce-fir
Kullarna N	62°00'	16°33'	295-440	150	till	clear-cut
Buskbäcken	59°55'	15°20'	285-350	181	till	spruce-fir
V Dybäcken	59°55'	13°55'	360-420	30	till	spruce-fir

Table 2 –  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for sieved soil samples from the investigated catchments

Locality	Sample	Depth (m)	Size (mm)	$^{87}\text{Sr}/^{86}\text{Sr}$
V Dybäcken	HF-1	C-horiz	<0.125	0.76122
V Dybäcken	HF-2	C-horiz	<0.125	0.76424
V Dybäcken	HF-3	C-horiz	<0.125	0.76588
V Dybäcken	HF-4	C-horiz	<0.125	0.75837
V Dybäcken	HF-5	C-horiz	<0.125	0.75859
V Dybäcken	B-3	C-horiz	<0.125	0.76048
Svartberget	SB-1	C-horiz	<0.125	0.73933
Svartberget	SB-2	C-horiz	<0.125	0.74053
Svartberget	SB-4	C-horiz	<0.125	0.74262
Svartberget	SB-7	C-horiz	<0.125	0.73814
Svartberget	JC-3	C-horiz	<0.125	0.74094
Svartberget	KC-3	C-horiz	<0.125	0.73961
Kullarna	W 1-B	0.1-0.2	<0.125	0.75801
Kullarna	W 2-B	0.1-0.2	<0.125	0.75920
Kullarna	W 3-B	0.1-0.2	<0.125	0.75530
Kullarna	W 4-B	0.1-0.2	<0.125	0.75679
Kullarna	W 5-B	0.1-0.2	<0.125	0.75585
Kullarna	KN-5	0.8	<0.125	0.76246
Kullarna	KN-3	1.0	<0.125	0.76424
Kullarna	KÖ-1	0.40-0.50	<0.125	0.77188
Kullarna	KÖ-2	0.60-0.65	<0.125	0.76463
Kullarna	KÖ-8	1.0-2.0	<0.125	0.75955
Kullarna	KÖ-8	1.0-2.0	0.250-0.125	0.76751
Kullarna	KÖ-8	1.0-2.0	1.0-0.25	0.77938

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Table 3 –  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for runoff water, precipitation and snow from the investigated catchments

Locality	Sample	Date	Depth (m)	$^{87}\text{Sr}/^{86}\text{Sr}$
Svartberget	Runoff	85-03-28		0.74077
Svartberget	Runoff	85-05-02		0.73920
Svartberget	Runoff	85-05-13		0.73805
Svartberget	Runoff	85-05-31		0.73825
Svartberget	Runoff	85-06-14		0.73895
Svartberget	Runoff	85-07-01		0.74059
Svartberget	Runoff	85-07-15		0.74202
Svartberget	Runoff	85-07-31		0.74192
Svartberget	Runoff	85-08-21		0.73962
Svartberget	Runoff	85-08-30		0.73860
		Duplicate		0.73872
Svartberget	Runoff	85-09-17		0.73817
		Duplicate		0.73822
Svartberget	Runoff	85-10-02		0.73896
		Duplicate		0.73901
Svartberget	Runoff	85-10-16		0.73808
		Duplicate		0.73811
Svartberget	Runoff	85-10-31		0.73930
Svartberget	Runoff	85-01-02		0.74243
Svartberget	Snow	85-04-02		0.71724
Svartberget	Snow	85-04-02		0.71846
Svartberget	Precip.	85-2/4-13/5		0.71608
		Duplicate		0.71630
Svartberget	Precip.	85-07-22		0.71555
Svartberget	Precip.	85-08-12		0.71197
Svartberget	Precip.	86-10-??		0.71492
Kullarna Ö	Runoff	85-03-23		0.75163
Kullarna Ö	Runoff	85-04-06		0.75156
Kullarna Ö	Runoff	85-04-12		0.75196
Kullarna Ö	Runoff	85-08-02		0.75233
Kullarna N	Runoff	85-03-23		0.75283
Kullarna N	Runoff	85-04-06		0.75324
Kullarna N	Runoff	85-04-12		0.75289
Kullarna N	Runoff	85-08-02		0.75045
Kullarna N	Snow	85-03-31		0.73290
Kullarna N	Snow	85-03-31		0.71488
Kullarna N	Snow	85-03-31		0.71657

continued

Table 3 continued

Locality	Sample	Date	Depth (m)	$^{87}\text{Sr}/^{86}\text{Sr}$
Buskbäcken W	Soilw.	85-10-27	0.05	0.74176
Buskbäcken W	Soilw.	85-10-27	0.10	0.73849
Buskbäcken W	Soilw.	85-10-27	0.30	0.73709
Buskbäcken W	Soilw.	85-10-27	0.75	0.73742
Buskbäcken W	Groundw.	85-10-21	1-2	0.73707
Buskbäcken W	Soilw.		0.05	0.74112
Buskbäcken W	Soilw.		0.10	0.73895
Buskbäcken W	Soilw.		0.30	0.73782
Buskbäcken W	Soilw.		0.75	0.73855
Buskbäcken W	Groundw.		1-2	0.73242
Buskbäcken SE	Soilw.		0.05	0.75445
Buskbäcken SE	Soilw.		0.10	0.74020
Buskbäcken SE	Soilw.		0.30	0.73163
Buskbäcken SE	Soilw.		0.60	0.73163
Buskbäcken SE	ZON-DEC			0.74200
Buskbäcken SE	Throughfall			0.73454
Buskbäcken	Sphagnum			0.72102
Buskbäcken	Above w.			0.73640
Buskbäcken	Under w.			0.73718
Buskbäcken	Soilw.		0.0-0.1	0.73643
Buskbäcken	Soilw.		0.1-0.2	0.73613
Buskbäcken	Soilw.		0.2-0.3	0.73470
V Dybäcken	Runoff	85-05-02		0.73313
V Dybäcken	Runoff	85-08-11		0.73274
V Dybäcken	Runoff	85-11-06		0.73212
V Dybäcken	Runoff	85-09-08		0.73264
V Dybäcken	Runoff	85-10-01		0.73282
V Dybäcken	Runoff	85-12-10		0.73262
V Dybäcken	Runoff	86-01-13		0.73330
V Dybäcken	Runoff	86-03-18		0.73361
V Dybäcken	Runoff	86-04-27		0.73288
V Dybäcken	Snow	85-10/12-28/12		0.71629
V Dybäcken	Snow	86-8/4-26/4		0.71042
V Dybäcken	Precip.	85-4/8-8/9		0.71855
V Dybäcken	Precip.	85-10/9-30/9		0.71462
V Dybäcken	Precip.	85-1/10-5/11		0.71271
V Dybäcken	Precip.	85-6/11-10/12		0.71623

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Table 4 - Grain size dependency for the strontium ratio between silt (Kullarna) and soil (Buskbäcken)

Sample	Size (mm)	Weight (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
Kullarna	1.0-0.25	29	0.7794
Kullarna	0.250-0.125	19	0.7675
Kullarna	<0.125	40	0.7596
Buskbäcken	>0.500		0.7733
Buskbäcken	0.500-0.125		0.7786
Buskbäcken	<0.125		0.7713

Sweden and central Europe and to some extent also the sea.

Concerning the strontium isotope determinations of soils we have generally used the <0.125 mm fraction of the tills. In sandy soils we have included also the 0.125-0.250 mm fraction as well. In general this represents more than 50 % of the till as a whole and probably more than 90 % of the total surface area. To illustrate the grain size dependency of the strontium isotope ratio a soil sample from Kullarna (Övre + Nedre) was split up in three fractions, and isotope determinations were performed on each one of them. The same procedure was done for a sample from Buskbäcken. While the sample from Buskbäcken was a true till soil, the sample from Kullarna was a water-reworked soil, essentially a silt. Results are presented in Table 4.

The silt sample (Kullarna Övre) shows a considerable fractionation which may be due to differential crushing and transportation of the minerals during the sediment formation. The value of the finer fraction (<0.125 mm) is considered to be the most representative one from the weathering point of view. The till sample from Buskbäcken shows quite small variations from one size fraction to the next size fraction (Table 4).

Åberg and Jacks (1989) performed twelve Sr isotope determinations of runoff water from the Buskbäcken site. The samples were taken during different flow rates during the year. Plotted in a diagram the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios showed a strong flow rate dependency (Åberg and Jacks 1989).

The observed discharge dependency must be due to a flow path located nearer to the surface during high flood occurrences and a larger fraction of atmospheric components in the water. A similar conclusion was drawn by Wickman and Åberg (1987) from a Sr isotope investigation of some source-lakes near the Buskbäcken catchment. In the other areas there are fewer runoff waters analyzed, but briefly the discharge dependency seems to be less. This is probably due to the impermeable subsoil in Buskbäcken (Lundin 1982). We believe that we can use the presently available runoff isotope data and obtain a reasonable result for the atmospheric-Sr to weathering-Sr ratio.

Table 5 – Atmospheric strontium in runoff water calculated from a compilation of the 87-Sr/86-Sr values in Tables 2 and 3

Catchment	Snow/Rain	Soil#	Runoff	Atmospheric strontium (%)
Svartberget	0.7168	0.7402	0.7398	2
Kullarna Ö	0.7156	0.7609	0.7519	20
Kullarna N	0.7156	0.7609	0.7521	20
Buskbäcken	0.7140	0.7720	0.7360	62
V Dybäcken	0.7140	0.7615	0.7371	53

#Represents the C-horizon of the podsolc soils.

Table 6 – Deposition and weathering rates of calcium calculated from strontium values

Catchment	Prec. (mm)	Ca2+ (mg/l)	Wet.dep. (kg/ha y)	Wet + Dry (kg/ha y)	Weathering (kg/ha y)
Svartberget	540	0.10	0.54	0.6	–
Kullarna	750	0.14	1.05	1.2	4.8
Buskbäcken	900	0.20	1.8	2.2	1.5
V Dybäcken	1000	0.20	2.0	2.4	2.1

## Weathering Rates

The strontium isotope ratios in runoff water falls somewhere between the precipitation values and the mineral soil values. This makes it possible to calculate the fraction of atmospheric strontium in runoff water (Table 5).

The figure for atmospheric strontium in Svartberget is only approximate as the ratios of soil and runoff lie so close to each other. It is however clear that the runoff is almost completely devoid of atmospheric strontium. The figures in Table 5 are based on about 5-6 samples collected from each compartment. In general the spread is small. As is already mentioned a certain discharge dependancy is observed for the Buskbäcken catchment (Åberg and Jacks 1989). For Buskbäcken the isotope ratio for runoff water has been discharge weighted. So far we do not consider that warranted for the other catchments.

The fractionations observed between calcium and strontium are small (Åberg and Jacks 1989) and this justifies drawing a parallel between the two elements. Under certain circumstances Sr does not correspond to calcium completely but deviates to some degree (Wadleigh *et al.* 1985). However, after consideration and for reasons of simplicity, we have assumed here that strontium and calcium correspond totally. Table 6 shows the deposition and weathering rates of calcium based



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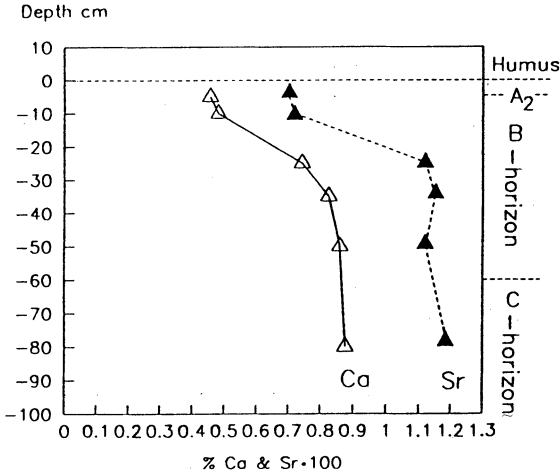


Fig. 2.  
Variation of Ca and Sr concentrations with depth in sieved soil samples (<0.125 mm) from Buskbäcken catchment.

on the assumption that calcium exhibits the same ratio of deposition to weathering as does strontium. But for Svartberget, the figure seems unrealistically high. To arrive at a reasonable rate more analyses of soil and runoff water are required.

The difference in weathering rate between Kullarna (Övre + Nedre) on the one hand Buskbäcken and V Dybäcken on the other is reasonable in view of their mineralogies. The former catchments have abundant plagioclases and a considerable amount of dark minerals while the latter two are characterized by soils rich in potassium feldspars. The figure arrived at for Buskbäcken (less than 2 kg Ca/ha year) can be compared with the historical weathering rate deduced from the depletion of calcium in a soil profile (Fig. 2). It is considered that the area was deglaciated 9,700 years BC (Lundqvist *et al.* 1961). Zirconium was used to calculate total loss of substance and it was considered that no volume changes have taken place. The mean weathering rate from 9,700 years BC until today was found to be about 2.2 kg Ca/ha year. The present day rate may be both higher and lower. Lower because of the fact that easily weatherable minerals are already lost in the soil profile. Higher due to the increase in atmospheric load and due to the development of vegetation.

### Calcium Budgets

Weathering and atmospheric deposition constitute inputs into the catchments while runoff and nutrient uptake by trees constitute losses. The inputs are calculated in the previous section. Only the biomass accumulation in stemwood and bark is considered to be a permanent loss from the catchment as with timber harvesting these are removed from the catchment. In general forestry practice branches with twigs and needles are left on the ground to decay.

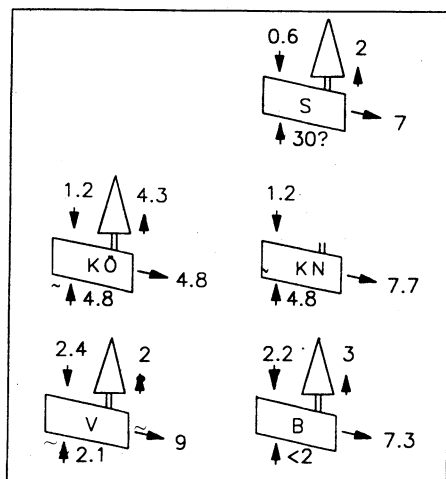


Fig. 3.  
Tentative Ca-budgets for Svartberget (S),  
Kullarna Övre (KÖ), Kullarna Nedre  
(KN), V Dybäcken (V) and Buskbäcken  
catchments (B).

The uptake of calcium for Svartberget and Buskbäcken are calculated from figures on annual stem volume increment (Grip 1982). The calcium uptake for Kullarna Övre catchment has been deduced from the figures presented by Rosén (1982). Out of a total uptake of 4.3 kg Ca/ha year a little more than 50% goes to the stem and bark, thus 2.2 kg Ca/ha year. The figure for V Dybäcken is based on the observation that the stand is thinner than that in Buskbäcken and thus should have less calcium uptake.

The export of calcium is based on records of varying length. The figure for Svartberget is based on two year's measurements when the precipitation was very close to the long term mean value (Degermark 1985). The Kullarna Övre series represents nine years of measurements (Rosén 1987). Kullarna Nedre has been clearcut for five years (Rosén 1987). The export from Buskbäcken is based on figures for 1979-1985 (Åberg and Jacks 1989). The transport from V Dybäcken finally is based on two years of observations.

The budgets are compiled in Fig. 3. The Svartberget catchment may be in steady state with weathering equalizing losses. The catchment is not very acidic as bicarbonate alkalinity occurs between the high flow events. Further soil and runoff water analyses are needed for determining the weathering rate with certainty.

The Kullarna catchments (Övre + Nedre) seem to be close to steady state. The reduced nutrient uptake in the clearcut catchment is transferred to runoff. The transport from the clearcut area is to some extent enhanced by nitrification (Rosén 1987).

Buskbäcken and V Dybäcken show clearly negative budgets. The main losses are via runoff. A good indicator to the processes behind the mobilization of an element is the anion composition in water (Dickson 1979). The mean anion composition for the two catchments is given in Table 7.

Sulphate is the dominant anion. In the Buskbäcken catchment the wet deposi-

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Table 7 – The mean anion composition for two catchments

Catchment	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	HCO <sub>3</sub> <sup>-</sup>	Org. anions (µeq/l)
Buskbäcken	160	75	6	0	70
V Dybäcken	120	50	1	0	50

tion has been calculated as 2.2. kg Ca/ha year as a mean for the period 1979-1985 (Åberg and Jacks 1989). The export by runoff of SO<sub>4</sub>-S was for the same period 7.3 kg Ca/ha year. Thus taking into account the dry deposition there is probably some accumulation of sulphur in the catchment. It is thus obvious that the atmospheric deposition plays a major role in increasing the losses.

### Conclusions

Using strontium behaviour as a model for calcium gives reasonable estimates of weathering rate in four out of five catchments. The calcium weathering rate was found to vary between 1.5 and 4.8 kg Ca/ha year. The weathering rate is correlated to the mineralogy in the sense that the lower figures are found in catchments with soils derived from acid granitoids (lower in Ca-content) while the higher figure is valid for soils derived from intermediate granitoids (higher in Ca-content).

Two of the catchments, those with the lowest weathering rates, have clearly negative calcium budgets. The main loss of calcium occurs via runoff with sulphate as the accompanying anion.

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