

THE USE OF NATURAL STRONTIUM ISOTOPES AS TRACERS IN ENVIRONMENTAL STUDIES

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Abstract. Naturally occurring isotopes of elements like strontium have proved to be good tools for tracing the past and for monitoring of processes in the present. The spread and variation in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios make Sr isotopes a powerful tool when it comes to detecting trends in the soil-vegetation system. There is also a great potential in combining different parameters like soil, water, biological material and isotopes for detecting environmental changes over short as well as long time periods. Sampling of the past is a difficult task but biological material, in combination with inorganic material, proves advantageous as environmental archives. There is also the possibility of using museum collections as environmental historic archives. This paper discusses the potential of using the natural $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as a tracer for environmental studies. The results presented point to an ongoing impoverishment of the environment and show that the temporal trend in Sr isotope composition for different media is similar despite material and location of test area.

Introduction

Environmental changes due to human activities cause increasing disturbances in nature and pose a threat to future life on earth. Natural conditions and natural development in relation to manmade processes have thus to be clarified since the human influence on nature more and more turns out to be a question of doubtful impact.

Accordingly, knowledge about chemical-biological processes in the environment is a prerequisite for a continued use of natural resources. The earth in itself can be seen as a gigantic laboratory and archive in which a number of biological, chemical and physical processes take place continuously, and in which the results of the past are recorded for posterity.

The Rb-Sr method is common for the dating of rocks, especially Precambrian, due to the long half-life of ^{87}Rb , but the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio can also be used as a petrogenetic tracer in granitic and gneissic rocks in studies of their source, element mobility in cracks and fissures, and the interaction between circulating fluids and their host rock. Proterozoic rocks, and soils derived from them, are enriched in ^{87}Sr , formed by radiogenic decay of ^{87}Rb . In old rocks, especially those rich in K, the ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ is high, and spreads out over a large interval, since Rb behaves similarly to K in many geologic processes.

Strontium is a common trace element in sea water with a concentration of about 8 mg L^{-1} . The isotopic composition of Sr in the oceans ($^{87}\text{Sr}/^{86}\text{Sr}=0.7092$) results from a mixture of Sr from young volcanics (ca. 0.703), marine carbonates

(ca. 0.708), and older continental crust (ca. 0.720) (Faure, 1986). Comparing the residence time for Sr in the oceans (about 2×10^7 years) with the ocean water mixing rate (about 10^3 years) the Sr isotopic composition in the oceans can be seen as constant at a specific moment. The Sr concentration of rainwater, which has an isotopic composition similar to ocean water but can be influenced by continental dust sources (Biscaye, *et al.* 1974), is less than $1 \mu\text{gL}^{-1}$; stream water in Sweden has Sr concentrations of about $20\text{--}40 \mu\text{gL}^{-1}$.

Over the last decade there has been an increasing interest in the Sr technique as a tool in studies of rates of weathering or determination of soil exchangeable-cation loss, for example. Among the early works are those of Graustein and Armstrong (1983) and Gosz *et al.* (1983), while more recent studies have been reported by Miller *et al.* (1993) and Blum *et al.* (1993).

The aim of this compilation is to show the potential of using natural Sr isotope ratios as tracers for environmental studies. Analyses of water (precipitation, throughfall, runoff, soil water), soil (mineral matter) and biological material (trees, mussel-shells, bones) show that the use of the Sr isotope ratio ($^{87}\text{Sr}/^{86}\text{Sr}$) is a powerful tool in estimating and understanding environmental changes, especially in areas dominated by Proterozoic acid rocks such as Sweden.

The use of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for fingerprinting the local Sr-ratio is shown in Figure 1. A cod and a shark from the open ocean give the supposed value of about 0.7092 while a cod from the Baltic Sea gives a slightly higher value (Schmitz *et al.*, 1991). The Baltic sea contains brackish water and gets a continuous input of freshwater with a higher Sr-ratio from rivers surrounding it. A pike and a moose from Central Sweden give high Sr-ratios of about 0.735 which is in accordance with other analyses from this area which reflects the influence of the Proterozoic soil. The lowest value (0.706) is from a reindeer from Iceland, and is intermediate between grass growing on the volcanic soil with a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio between 0.703 and 0.704, and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of rainwater of about 0.709.

Our samples have been analysed on Finnigan MAT 261 mass spectrometers at the Laboratory for Isotope Geology, Sweden and Institutt for energiteknikk, Norway and on Micromass 54 E mass spectrometers at Scottish Universities Research and Reactor Centre (SURRC), Scotland and Commonwealth Scientific Industrial Research Organization (CSIRO), Australia.

Strontium in the Environment

WATER AND PRECIPITATION

In a study of annual variation in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in source lakes from Central Sweden, two localities were chosen situated in a bedrock consisting predominantly of approximately 1700 to 2000 Myr old granites and gneisses (Wickman and Åberg, 1987). The unconsolidated cover rocks are mainly till from the last glaciation, which

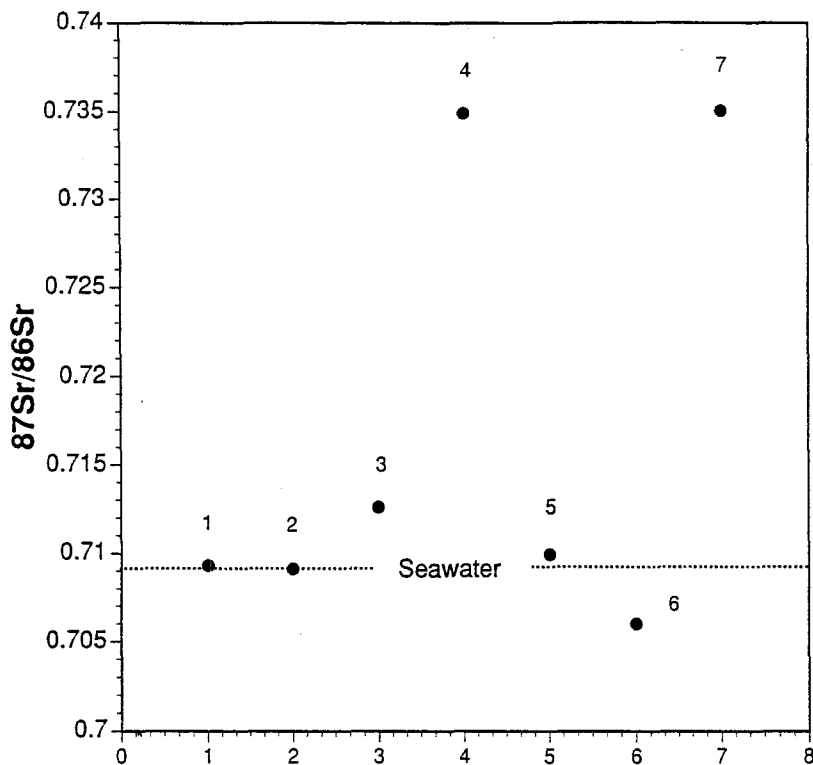


Fig. 1. Sr isotope ratios in different fish and mammal skeletons. 1. Cod from the North Sea, 2. Shark from Coast of Morocco, 3. Pike from lake in Southern Sweden, 4. Pike from lake in Central Sweden, 5. Cod from Southern Baltic Sea, 6. Reindeer from Iceland, 7. Moose from Central Sweden.

ended there about 10,000 years ago. These lakes were sampled monthly during one year in order to study seasonal variation in the isotope ratios (Figure 2). The lakes were covered by ice early in December 1982; the ice cleared by May 1, 1983. The ratios varied in a narrow range except during the spring thaw period when the ratio of the surface water was lowered by a contribution from melting snow. Large masses of snow with a Sr isotope ratio of 0.710 had accumulated during winter. At the clearing of ice the snow ratio had a sudden impact on the surface water, but this impact was short-lived due to the low Sr concentration in the snow less than $1\mu\text{gL}^{-1}$. Small simultaneous changes in the isotope ratios of the two lakes were observed in July and August. Since the isotope values were nearly constant most of the year and the distance between the lakes is only 2 km, it is natural to assume that a common cause for such seasonal variations is precipitation. Periods of more or less continuous rainy weather or episodic heavy rains, with a low Sr isotope ratio, depress the lake water isotope ratio, while periods of dryness may increase the lake water isotope ratios. Available data on the local weather in July-August are consistent with this interpretation.

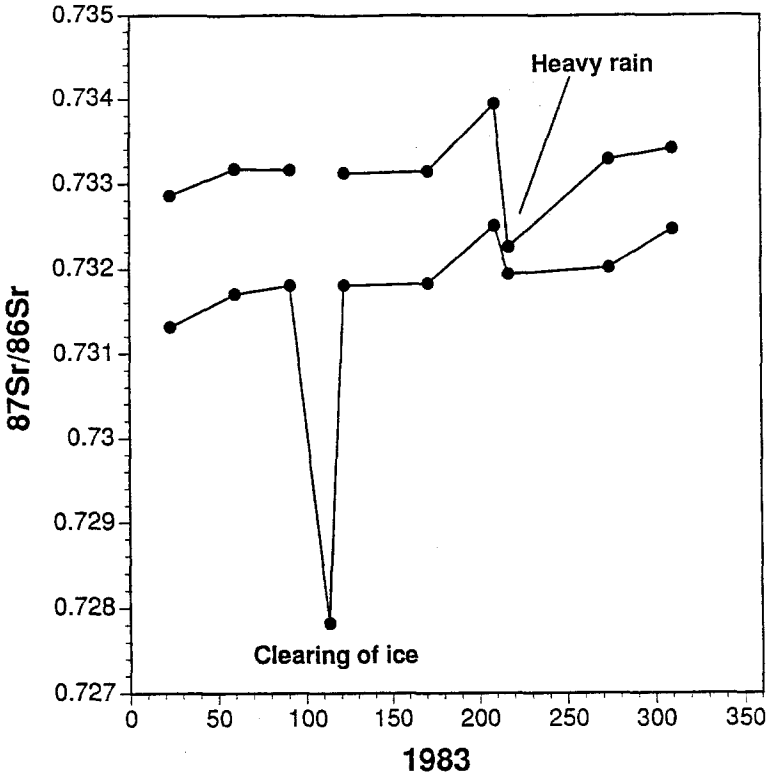


Fig. 2. The Sr isotope ratio in two fresh-water lakes varied in a narrow range over one year. During clearing of ice it decreased strongly due to the impact of snow with a low Sr isotope ratio. The increase and decrease in late summer was due to a long dry period followed by heavy rains.

When the value from April 24 (Figure 2) is disregarded the mean Sr isotope ratios of the lake waters are 0.732 and 0.733. These values correspond fairly well to those observed for the pike and moose from this area (Figure 1) and for water from streams feeding into the Bothnian Bay of the Baltic Sea. The variation in Sr isotope ratio, with respect to bedrock geology, suggests that an important influence on the ratios is the mineralogical composition of the Quaternary deposits, mainly till. The observed limited variations are therefore an expression of the fact that granitoid, Proterozoic rocks are common in this part of Central Sweden, and that mafic rocks and limestones are only minor constituents.

In another study (Åberg *et al.*, 1989) we found a tendency for an inverse relationship between the Sr isotope ratio and discharge for a stream in an area in Central Sweden (Figure 3). Abundant rainwater with a low Sr-ratio will have a greater impact on the runoff value at high discharge. The residence time for soil water will also decrease with increased precipitation and thus limit the time for interaction with the soil minerals.

Contrasting rock compositions in river basins, especially when combined with large age differences, result in greatly differing Sr isotope ratios for river waters.

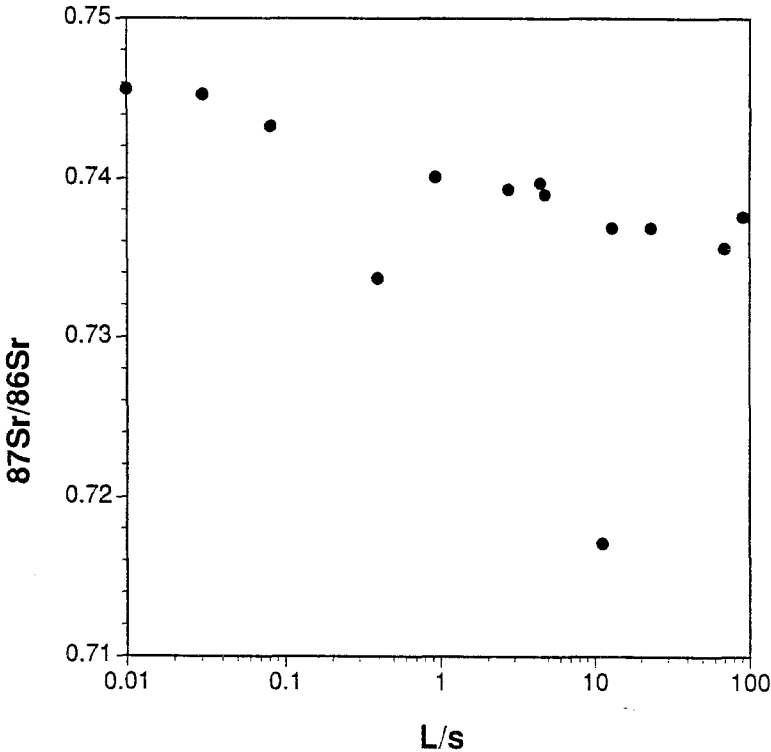


Fig. 3. Within a river there is an inverse relationship between Sr isotope ratios and discharge, i.e., high Sr isotope ratios are associated with low discharge and vice versa.

Wadleigh *et al.* (1985) discussed the Sr ratios of water from 39 major Canadian rivers with discharge rates similar to or larger than our largest streams.

Their Sr isotope ratios were clustered into three main groups. One group had ratios around 0.705 and represented rivers from the young basalt province on the Canadian west coast. A second group had ratios ranging from about the rain-sea water value, 0.709, up to about 0.720, and correspond to rivers in drainage areas where many kinds of rocks, including granitoids, limestones, etc. are mixed. A third group had ratios from about 0.725 to 0.740; these rivers come from old, Proterozoic-Archean granitic areas around Hudson Bay. Overall, the Canadian rivers have drained an area with great variations in rock types and ages, which explains the large spread of their values.

We analyzed samples from 45 rivers in Sweden and Finland (Åberg and Wickman, 1987). Our weighted average Sr isotope ratios were high (Figure 4), reflecting the dominance of granitic Proterozoic rocks in the region. Even though we cannot draw a direct parallel between our study and that by Wadleigh *et al.* (1985) because areas with young basaltic composition are lacking in the Baltic Shield, we can see that rivers draining bedrock of similar composition and age give similar results in

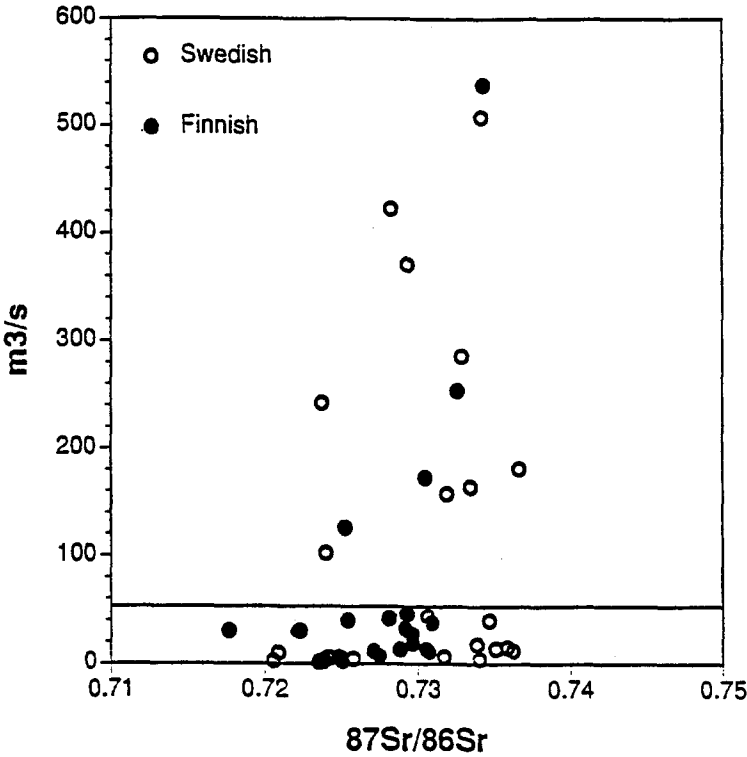


Fig. 4. Comparing rivers, a high discharge rate is associated with a smaller spread in Sr isotope ratios and a low discharge with a larger spread. That is, rare rocks have a larger impact in a smaller catchment.

both shields. Around the southern part of the Baltic, however, the impact of rocks from the Paleozoic-Mesozoic carbonate sedimentary basins can be seen in the low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the rivers draining these areas (Löfvendahl *et al.*, 1990).

Figure 4 shows discharge against isotope ratios. Rivers with a low discharge rate between ($1\text{--}50\text{ m}^3\text{ sec}^{-1}$) have Sr isotope ratios ranging from 0.718 to 0.736 (Åberg and Wickman, 1987). As discharge increases, the spread in Sr isotope ratios decreases and the mean Sr isotope ratio shifts toward a higher value, reflecting a greater influence by the acid Proterozoic rocks.

Why is the spread greater and why do low Sr isotope ratios mainly occur in streams with small discharge? One cause is the occurrence of rocks with low Sr isotope ratios like mafic rocks and limestones. If they form a considerable part of the bedrock and thus of the unconsolidated deposits, a lower Sr isotope ratio will result. The pattern that in the Baltic Bay region low Sr isotope ratios only exist for small streams, can therefore be interpreted as follows: the rocks are rare and of only local importance. That is, in a small drainage area they may, by chance, be important, but for a larger drainage area this situation is unlikely.

A study of major elements, stable isotopes and Sr isotope ratios in a West-East snow profile across the Caledonides, Central Scandinavia (Andersson *et al.*, 1990), showed an increase in Sr isotope ratios and a decrease in stable O isotope ratios with distance from the Atlantic Ocean (Figure 5). The highest peaks along the transect reach up to about 1600 m a.s.l. while the highest sampling point was at about 500 m a.s.l.

The easternmost sample had a low stable O (and H) isotope ratio and low concentrations of Na^+ and Cl^- ; the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (and the S concentration), however, reached its highest value along the transect. This sample is strongly influenced by anthropogenic pollution. A good candidate for the pollution is the wood pulp industry which is localised along the coast here. The factories are using bark and wood waste as fuel for energy production. Wood is also commonly used in the area for domestic heating.

In a study of the geochemical cycle of Sr results from Central Sweden yielded ratios around 0.71 for precipitation (Åberg and Jacks, 1985, 1987). Throughfall (below the canopy), with Sr isotope ratios on the order of 0.72–0.73, is a mixture of atmospherically transported Sr caught by vegetation and Sr from the mineral soil. Similar observations were made by Graustein and Armstrong (1983) who found that Sr in forests was derived mainly from two sources: weathering and dust from remote areas transported through the atmosphere. Further results (Åberg and Jacks 1985, 1987) showed that shallow ground water in a dug well and runoff water had ratios of around 0.73–0.74 and water from a fracture zone in a stream gave a value of 0.78. The rock in the fracture zone had a Sr isotope ratio of 0.85 to 0.90.

SOIL

Analyses of the Sr isotope ratio for soil samples indicate that weathering is related to the size of the mineral grains; smaller grains have a lower Sr isotope ratio than larger grains (Jacks *et al.*, 1989; Åberg *et al.*, 1990). An explanation may be that the smaller grains greater surface to volume ratio results in a stronger weathering ability which in turn may give rise to a preferential leaching. That is, ^{87}Rb has a different ion-radius and charge than its ^{87}Sr daughter. It may be that $^{87}\text{Sr}^+$ ions leave the Rb site more easily than the more common $^{86}\text{Sr}^+$ ion which is more tightly bonded in the lattice.

In depth profiles (Åberg *et al.*, 1990) of two soils Sr isotope ratios were higher in the uppermost layer and decreased downwards to around the 20 cm depth (Figure 6). Below 20 cm, the Sr isotope ratio stabilized and this trend continues to ground water level. Higher ratios in the upper soil depths are probably due to more K-rich and a depletion in more easily weathered Ca-rich minerals. Soil water samples follow the same depth trend as the soil samples (Åberg *et al.*, 1990) but may have lower Sr isotope ratios (Figure 6); the decrease in Sr isotope ratios appears here in the first 5 to 10 cm. Within an area sampled at random over some

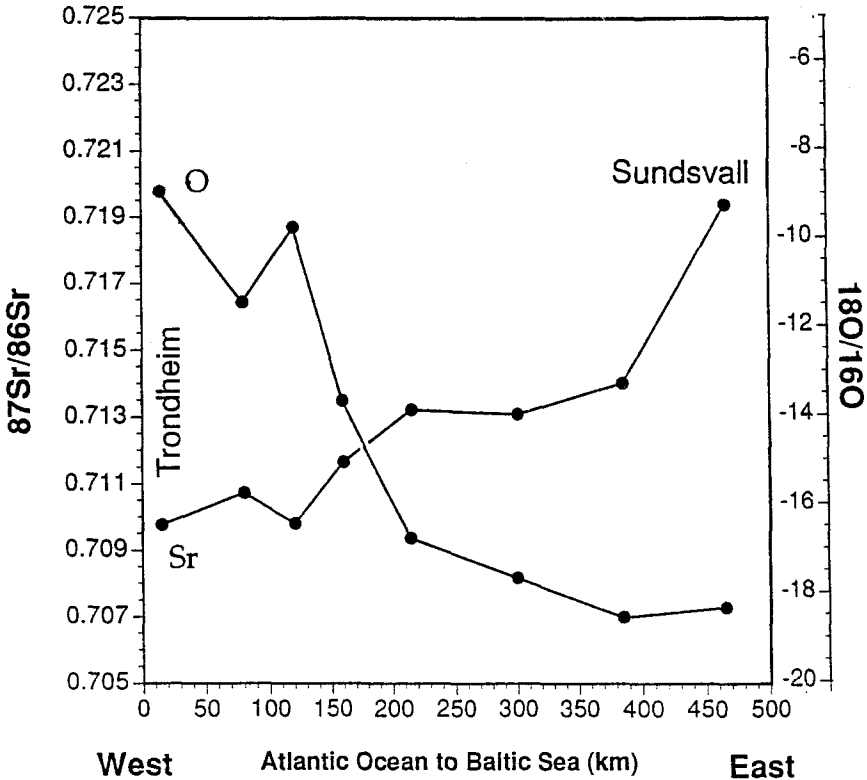


Fig. 5. Sr isotope and $\delta^{18}\text{O}$ ratios for precipitation are readily influenced by environmental factors with a marine (Trondheim) or anthropogenic signature (Sundsvall wood-pulp industry).

500 m², at the same depth, the soil is quite homogeneous having similar Sr isotope ratios (Figure 6).

The uppermost soil layer, -3 to 0 cm depth, consisting of humus (organic matter), is enriched in products that have passed up through the trees and then been deposited in the form of needles and twigs on the ground (Åberg *et al.*, 1990). Leaching of this soil layer with ammonium acetate gives a Sr isotope ratio similar to that of throughfall. Leaching of the upper mineral soil (0–15 cm), yields a slightly higher Sr isotope ratio than that recorded for both soil water and the previous humus layer. Exchange sites in the mineral soil layer consist of organic material that has been transported down from the humus layer and mixed with inorganic material (Åberg *et al.*, 1990). The Sr is largely adsorbed onto organic matter on the mineral grains.

BIOLOGY MATTER

Bark of spruce, pine and birch from two areas were analyzed for Sr isotopes (Figure 7). In an additional area, bark, wood from the trunk's outermost growth

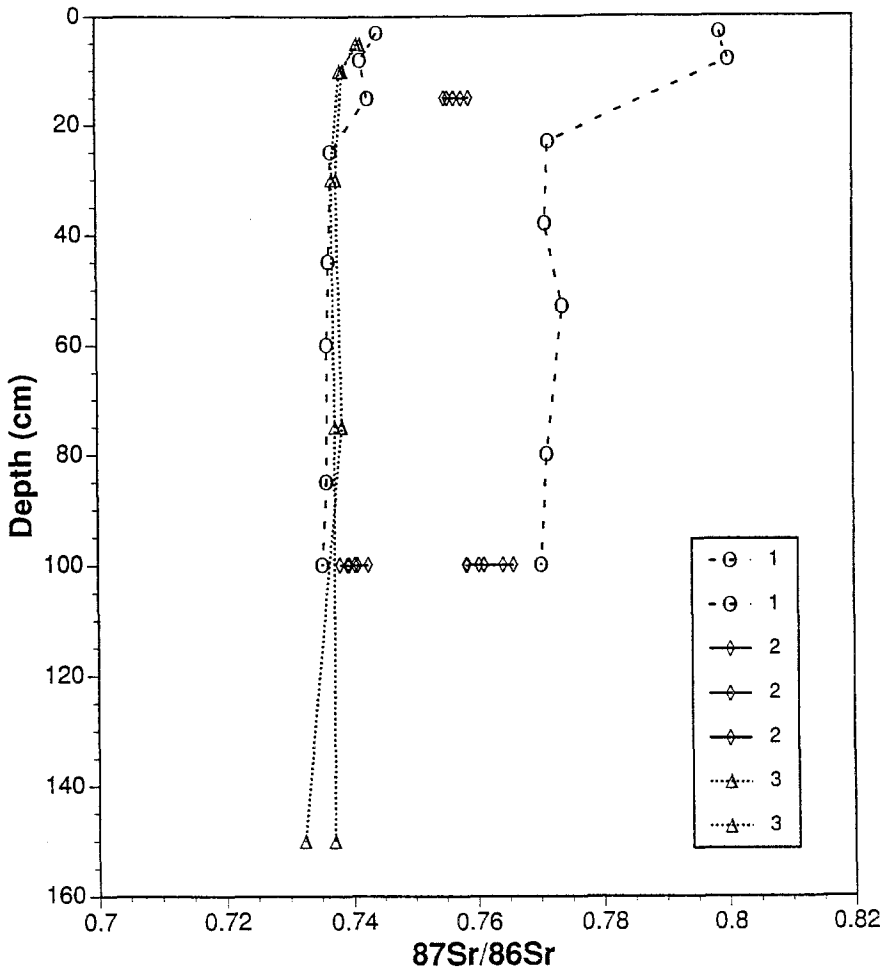


Fig. 6. (1) Variation in Sr isotope ratios in soil with depth for two localities. The uppermost part has lost Ca minerals (Sr-rich) due to weathering and is enriched in more resistant K-minerals (radiogenic ^{87}Sr -rich). (2) Homogeneity in Sr isotope ratios at three localities in soil at same depth within each locality. Each sampling area is about 500 m^2 ; grain size is $< 0.125 \text{ mm}$. (3) Variation in Sr isotope ratios with depth in soil water from one locality during 1985 and 1986.

layer and wood from the core were analyzed from spruce and pine (Åberg *et al.*, 1990). The age of the trees was about 40 yr. An advantage of analyses using tree material is the ability to determine the age of every growth ring. Over time, each growth ring becomes terminated and lignified, thereby preserving its Sr isotope ratio. Bark from spruce and pine gave similar values within each of the two areas; birch bark Sr isotope ratios were slightly displaced towards higher values, possibly due to the different root system of the birch. In the third area, spruce and pine trees had similar Sr isotope ratios for bark or for the trunk's outermost growth layer.

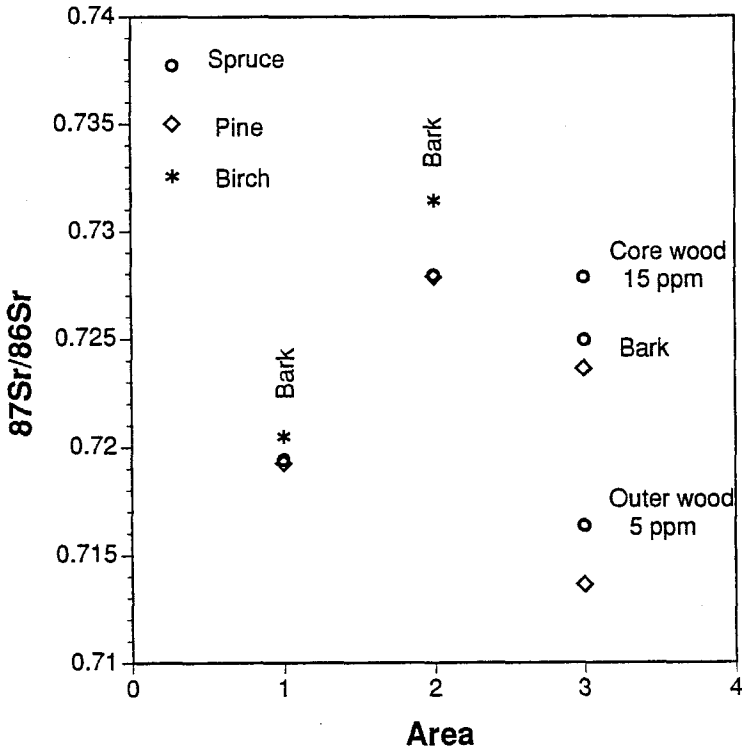


Fig. 7. Bark from spruce and pine give similar Sr isotope ratios for 2 areas. The birch ratios are slightly biased upwards, probably depending on different root systems. Core wood to outer wood shows decreasing Sr concentration and Sr isotope ratio for spruce from area 3.

Analyses of calcium in tree-rings from pine from two localities and spruce from one locality show a similar trend despite geographic location (Åberg *et al.*, 1990). After a high initial Ca content of about 1000 ppm there is a rapid decrease to 700–600 ppm. Then comes a slow decrease for about 25 years, followed by a rapid decrease to about 300 ppm in the late 1980's. The decrease in Ca content in the trees with time may have several explanations. For example, the tree when growing is impoverishing the soil and accumulating the nutrients in the trunkwood. Therefore, the new wood has less Ca available to take up from the soil. Alternatively, ongoing acidification of the soil will transport cations like Ca^{2+} out of the ecosystem and impoverish the soil. In this case the input of cations from natural weathering cannot keep up with the export out of the soil pool.

The decrease in Sr isotope ratio from 0.728 to 0.716 during 1942 (core wood) to 1985 (outer wood) is taken as reflecting the composition of the Sr isotopes entering the tree (Figure 7). Assuming the contribution of rainwater (Sr isotope ratio of ca. 0.71) to the soil to be constant over the years, a change toward a lower Sr isotope ratio in younger wood must result from a decreased contribution of Sr from the soil pool. This may happen if the export of cations from the soil pool out of the

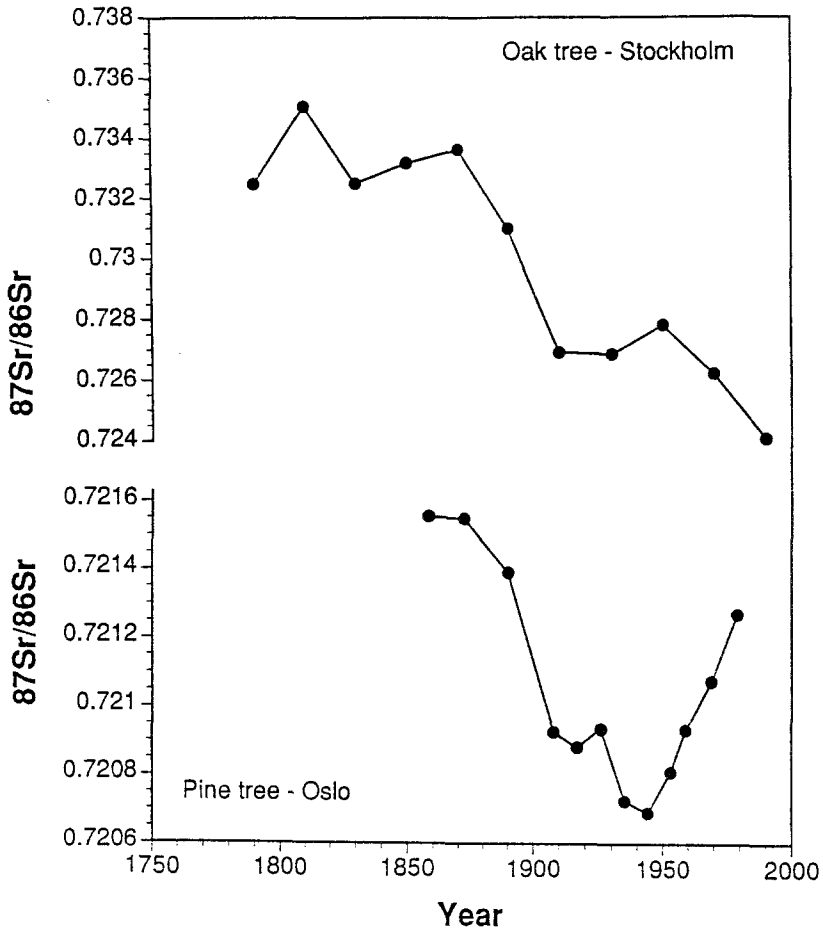


Fig. 8. Sr isotope ratios from an oak tree growing along a main road on the northern side of Stockholm and from a pine tree growing outside Oslo (note break and difference in scale for the Sr isotope ratio).

ecosystem is greater than the input from natural weathering. As a result, the Sr isotope ratio in the soil water taken up by the tree, moves towards that of rainwater. Isotopic analyses of an oak tree growing along a main road in the outskirts of Stockholm showed a decrease in Sr isotope ratio from 0.735 to 0.724 between 1790 and 1990 (Figure 8). A similar study of a pine from outside Oslo showed the same decrease in Sr isotope ratios between 1850 and 1950, but after that a steady increase until today (Figure 8, A. Råheim, pers. comm.).

In order to see if the decrease in Sr ratio over time was a general feature, we looked for another independent system and found the freshwater pearl mussel *Margaritifera* in Central Sweden. *Margaritifera* has a long life-span of over 100 years, making it a good environmental archive species.

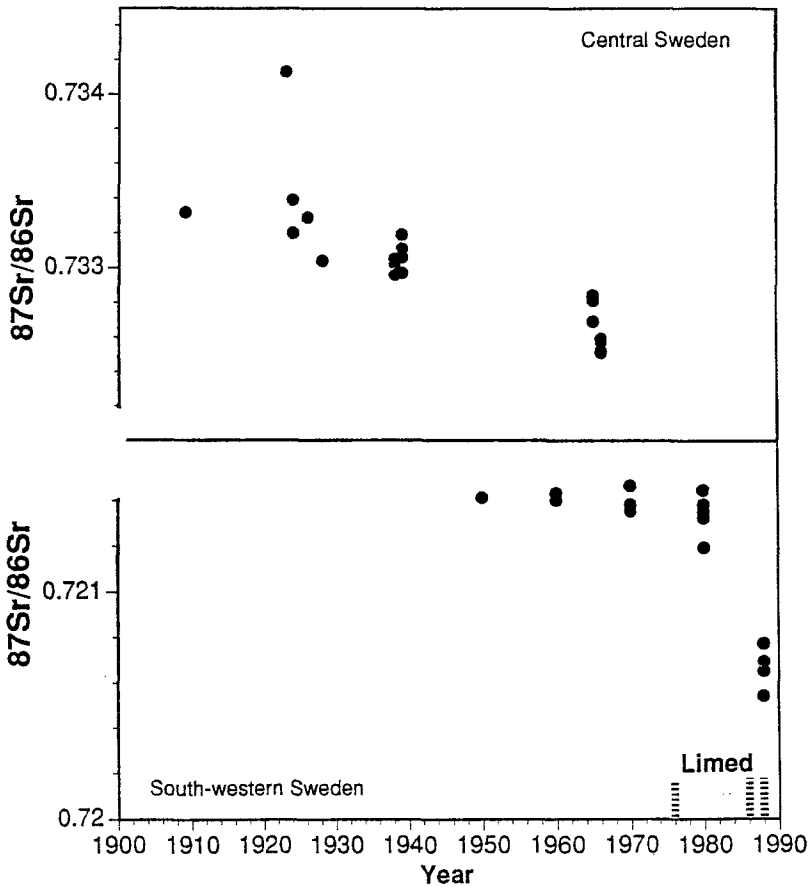


Fig. 9. Decreasing Sr isotope ratios in freshwater mussels from rivers in Central and South-western Sweden. The latter river was limed 1976, 86 and 88 with an obvious effect in the Sr isotope ratios (note break in Sr isotope ratio between the upper and lower figures).

The Sr isotope ratios from three investigated localities (Åberg et al., unpublished data) show a similar trend over time but in two different Sr intervals depending on geographic location. While the localities in Central Sweden are dominated by Proterozoic acid rocks, with Sr isotope ratios around 0.733, the locality in south-western Sweden is in an area consisting of younger rocks with Sr isotope ratios around 0.721.

Analyses of the mussel shells from the central Sweden sites show a distinct decrease in Sr isotope ratios from 1915 to 1990 (Figure 9). For the younger mussel from Slerebo there is a quite consistent decrease in Sr isotope ratios from 1945 to 1980 and then there is a sharp drop. This river, however, was limed 3 times around 1980 with a remarkable effect on the Sr isotope ratios. It is not known from where the limestone was quarried but a Sr isotope ratio of 0.708–0.710 would be quite reasonable for it. The result suggest that liming and uptake by biological tissue like

mussels may be traceable with stable Sr isotopes. The Sr isotope ratios obtained from the central Sweden sites are similar to the ratios reported for two source lakes, a pike and moose in the vicinity of these rivers (cf. Figures 1 and 2).

There is a distinct decrease in Sr isotope ratio in the *Margaritifera* shells with time, well outside the error limits. This is the same trend found when analyzing the Sr isotope ratio of tree rings in the vicinity. The latter showed a decrease both in element uptake and Sr isotope ratio by the Sr isotope ratio by the tree; the Sr concentration had decreased from about $15 \mu\text{g g}^{-1}$ in 1942 to about $5 \mu\text{g g}^{-1}$ in 1985, and the Sr isotope ratio decreased from 0.728 to 0.716 (Figure 7). Unlike trees, the mussels are not impoverishing the soil and accumulating the nutrients in the stemwood but mirror the composition of the environment they are living in.

A study by Falkengren-Grerup and Tyler (1992) on soils from the C-horizon in southern Sweden, originally sampled in 1947–52 and resampled 1988, showed a marked decrease in element concentration. The conclusion was that the decrease of both pH and exchangeable base cation pools indicate that the weathering rate of silicates does not keep pace with the accumulation of mineral nutrients in the biomass and the higher leaching rate caused by increased activities in the soil solution.

The results of Carell *et al.* (1987) for *Margaritifera* shells, from the Tansån river in central Sweden, point to similar conclusions. Elements like Au, Ag, Fe and Co clearly decrease in concentration with time. The reason, according to Carell *et al.*, is that increasing acidity might suppress the chemical/biological availability of Au and Ag and also the uptake of Fe and Co. They also reported an increase in Mn and S, which is in support of an 'ongoing acidification' hypothesis.

Summary

The results of the Sr isotope analyses point to a change over time in the environment and show that the compositional trend for different media is similar despite location of the test area. The spread and variation in the Sr isotope ratios thus makes Sr isotopes a powerful tool for detecting trends of base cation depletion in the soil vegetation system and especially together with other sensitive analytical methods. There is obviously a great potential in combining different parameters like soil, water, biological material and isotopes for the study of detecting environmental changes over shorter as well as longer time periods.

Since sampling of the past is a difficult task in many disciplines, biological material in combination with inorganic material, offers a great advantage as environmental archives. They make it possible to study any area in any place over a certain time interval. Trends in environmental changes can be studied also using museum collections.

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