An Investigation of Trace Elements in Marine and Lacustrine Deposits by Means of a Neutron Activation Method

O. Landström, K. Samsahl and C-G. Wenner



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ABSTRACT

A neutron activation method for multi-elemental analysis was applied to argillaceous deposits from the Viskan Valley on the Swedish West Coast, an area in which transgressions and regressions of the sea have occurred during the postglacial time. These changes between fresh-water and marine stages are well known from earlier geologic investigations. Analysis could therefore be performed of samples of known marine (brackish) and lacustrine origin.

In a preliminary test on four samples, taken from two marine layers and two lacustrine layers, altogether 30 elements were quantitatively determined, namely As, Br, Ca, Ce, Co, Cr, Cs, Eu, Fe, Ga, Hf, Hg, La, Lu, Mn, Na, Rb, Sb, Sc, Se, Sm, Sr, Ta, Tb, Th, U, W, Yb, Zn and Zr. Several elements showed a difference in concentration between the marine and the fresh-water layers. Br, Hg, Mn, Se, As and Sb were more thoroughly investigated in three boreholes from different sections of the valley.

Br and Se had high means in the most pronounced marine samples of the fiord deposits. Moreover, Mn showed a fourfold increase just in the transition stage between the older sea fiord and the older lage stage.

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1. INTRODUCTION

In the present work a neutron activation method, suitable for multielemental analysis, was adopted to lithologic material. The method was also applied in a preliminary study of the use of trace elements in distinguishing marine and lacustrine deposits in the Viskan Valley on the Swedish West Coast.

An extensive investigation of the different strata in this area was carried out by Professor Lennart von Post during the years 1932-1944 with the aim of making a graph of the shore fluctuations [1, 1938]. The material from this investigation seemed to be very suitable for the present test. The Viskan Valley represents a limited area, where the mineral constituent of the sediments comes from geologically homogeneous surroundings. During a rather short time, less than 8,000 years, marine and fresh-water conditions have alternated several times. These changes are well defined as they have been based on paleontologic evidence in many sections of the valley, and represent movements of the shore well known in the Quaternary history of the Viskan Valley.

Attempts have been made to use B as a salinity indicator in two sequences of strata from the Viskan Valley [2, 1945]. In one borehole the B content of the argillaceous sediment seems to reveal changes in the salinity of the deposition environment. In another borehole, where no changes in salinity are to be expected, the variations in the B content from level to level can be explained by the appearance of layers of sand and silt in the argillaceous deposit. Since these very first investigations the understanding of boron as a salinity indicator has been discussed by numerous geochemists [3, 1963].

In regions other than the Viskan Valley attempts have been made to differentiate marine and fresh-water sediments by means of trace elements. In Pennsylvanian sediments from the Appalachian Basin the concentrations of B, Li, F, Sr, S and Mn are higher in marine shales, while Ga and Cr are higher in the fresh-water shales of the investigated basin [4, 1959]. To avoid local influences, samples also have been collected and examined from geographically different regions [5, 1963]. In 66 samples from marine and fresh-water argillaceous sediments the elements B, Co, Cr, Cu, Ga, Ni, Pb, V and Zn were determined by spectrographic methods. B, Cr, Cu, Ga, Ni and V were found to be more abundant in marine than in fresh-water sediments. The most marked difference was found in B and V. No major differences in concentrations distinguished ancient and recent sediments from the same area though the 33 ancient rock samples represented formations from Ordovician to Pliocene.

2. SAMPLING LOCALITIES

In order to understand the sediments investigated, a short review is given of the geological history of the area. A more detailed description of the sediments will follow in connection with the results in Chapter No. 4.

After the Quaternary ice-sheet had melted from the Viskan Valley on the Swedish West Coast, a fiord of salt water stretched 60 km from the present coast line at Varberg to Seglora and Kinnarumma. As a result of land uplift the shore moved down towards the present coastline. When the sea-level passed a threshold at Veddige, situated 15 km from the sea, a lake was isolated in the Viskan Valley. On the marine beds of the sea fiord, fresh-water layers were deposited, but later the sealevel rose above the threshold at Veddige. The sea then returned to the lake and spread over the fresh-water sediments of the lake, and over terrestrial soils up towards Berghem about 40 km from the present coast at Varberg. After this great post-glacial transgresseion, which created a younger sea-fiord with new marine layers, the sea-level retreated past the threshold at Veddige, and another longish lake was isolated in the Viskan Valley, the present Lake Veselången. In the sequences of strata of the Viskan Valley there were, consequently, two marine stages (the older seafiord and the younger one) and two freshwater stages (the ancient lake and the present lake).

The series of samples which have now been tested are Nos. 629, 1511 and 16. They arise from three different sections of the Viskan Valley (fig. 1) situated about 20, 30 and 35 km from the present coastline. In the two sections first mentioned, representing mainly sediments in deep water, the strata are, in spite of different degrees of salinity, similar as regards the size of mineral particles and the content of organic materials. The section with bore-hole 16, on the other hand, represents a part of the Viskan Valley located near the highest limit of the post-glacial transgression during the younger sea-fiord stage. At the upper end of this fiord, where the post-glacial Viskan River discharges its waters, the water was up to a few meter deep. The result was a delta in the shallow water and swamps on the flood plain above the water level.

3. ANALYTIC WORK

3.1. Sampling and preparation

A large Hiller auger was used in the borings and samples 2x1 cm were cut out with a spatula from the inner part of the core after removing the superficial part. From the spatula the sample was packed around with cellophane. As the aim was to make a microscopic investigation of the samples, they were taken with great care and accuracy. The analytic results have hitherto not shown any signs of contamination.

For the activation analysis, about 15 mg of the samples were weighed, in polyethen ampoules for irradiation in reactor R1 in Stockholm, in quartz ampoules for irradiation in reactor R2 at Studsvik, Undried samples were used for the analysis and the dry weight (at 105° C) was determined using other parts of the samples.

3.2. Instrumental analysis

A preliminary investigation showed that at irradiation times of the order of 1 min, 10 min, 10 hours and 4 days respectively, relatively strong activities from some isotopes dominated the gammaradiation from the samples, namely Al-28 (2.3 min) Mn-56 (2.58 hours), Na-24 (15 hours), Fe-59 (45.1 days) and Sc-46 (85 days). Thus many nuclides with low activity were inaccessible with instrumental analysis. Accordingly, some form of radiochemical separation was found necessary and in the following section the application of a chemical separation method to the samples will be described. Using this procedure, elements with a half-life of the order of some hours could not be analyzed, and as Mn could be determined very easily by instrumental analysis, this was done in a first step.

For this the samples were irradiated in reactor R1 for about 3-5 min. in a flux of 6×10^{11} n/cm² sec. together with a Mn-standard. After the decay of A1-28 and Mg-27 the 0.84 MeV photopeak of Mn-56 was measured by gamma-ray spectrometry.

3.3. Chemical separation method

After the determination of Mn, the samples were re-irradiated in reactor R2 for about 3 days in a flux of $2 \times 10^{13} \text{ n/cm}^2$ sec. (the samples from borehole 1511 and 16, which were analyzed only for As, Br, Sb and Se, were irradiated in reactor R1). After 1 - 2 days decay, the radioactive constituents of the samples were separated into 20 groups with a chemical group separation method, shown in fig. 2. The method, consisting mainly in ion-exchange without carrier addition, is in principle the same as a routine procedure originally developed and used in neutron activation analysis of biological material [6, 1965]. However, the introduction of a few additional steps in the procedure were found necessary in the case of argillaceous samples.

One of these steps consists of the removal of silicic acid from the sample solutions by vaporizing SiF_4 from a mixture of H_2SO_4 and HF in the following way. After the initial division of the radioactive constituents of the sample into two main groups by means of distillation with H_2O_2 and HBr[7, 1962], the mixture of sulphuric and silicic acid remaining in the distillation flask was transferred to a platinum dish. 5 ml of conc. HF was then added and the mixture taken to copious fumes of SO_3 at low heat on a hot plate. The vaporization procedure was repeated 6 times in all before a H_2SO_4 solution free from silica was obtained. After the subsequent dilution of the sample the precipitated metallic gold was filtered on a G-4 glass filter disc.

Additional washing steps are applied to some of the ion exchange columns and precipitates in the series. These steps, which remove excessive amounts of mainly Sc-46 activity, were performed as follows (fig. 2). Groups 11, 12 and 13 were coupled in series and given an extra wash with 100 ml 6 n HCl, which was continuously passed through the columns at a rate of 4 ml/min.

The precipitate obtained in group 15 was decontaminated by an extra continuous washing with 50 ml 0.3 N HCl at a rate of 4 ml/min, whereas the samples of group 4 were cleaned solely with 30 ml 0.5 N HCl in a similar way.

3.4. Measurement of the radioactivity

The identification of the separated radioactive nuclides was performed by means of gamma-ray spectrometry, using a $3'' \times 3''$ well-type NaI detector and a 512-channel analyzer. Diagrams 1-18 show typical spectra of the different groups, recorded in connection with the analysis of a clay sample which had been irradiated 3 days in a flux of 2×10^{13} n/cm² sec. The spectra of groups 4, 10 and 20 have been recorded with the resin placed in the well of the crystal; the other resins were measured close to the detector. Groups 6 and 7 were mixed and measured together. The activity of most of the nuclides was sufficient to allow counting times of 1-20 minutes.

Table 1 lists those elements which have been quantitatively determined or qualitatively observed and also the nuclides on which these determinations have been based.

3.5. Calculation of weight concentrations

The concentration of each element was calculated by comparing their activity with that of simultaneously irradiated standards of known amounts. These standards were measured at a distance of 15 cm from the detector and the counting rates thus obtained were multiplied by predetermined correction factors in order to be valid for those geometrical conditions in which the different resins were measured. The correction factors also include corrections for losses in the chemical processes and were determined experimentally as described earlier [6, 1965].

The values given in table 2 have been corrected when necessary for errors due to fission products of uranium as well as other inter-

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fering reactions. The correction for the activity introduced by fission products has been calculated in a similar way as described by Loveridge et al. [8, 1959].

3.6. Discussion of the chemical separations

The reliability of decomposing the samples with a mixture of sulfuric acid and hydrogen peroxide prior to the distillation procedure was checked as follows:

Irradiated samples were dissolved in a few ml of water after fusion with a mixture of Na_2O_2 and NaOH at 600°C. The solution, containing added carriers in an amount corresponding to 5 mg each of As, Br, Hg, Sb och Se, was strongly acidified with H_2SO_4 and the elements were finally distilled [7, 1962]. The activity in the distillate was, except Hg, in full agreement with those obtained with the sulfuric acid method. Regarding Hg, lower values were obtained after a Na_2O_2 attack, probably owing to volatilization during the fusion process. However, the main reason for using a mixture of H_2SO_4 and H_2O_2 as a decomposing agent in the present work was to keep the salt concentration down in the solutions used for the subsequent ion-exchange separation steps.

To determine whether elements other than silicium were vaporized, completely or in part, during the hydrofluoric acid treatment, tests were carried out as follows:

The platinum dish containing the radioactive sample solution after the previous distillation procedure was covered with a glass funnel. The funnel was connected by a short polyethene tube to a washing bottle containing cold 6 N NaOH. A rapid stream of air was drawn through the system during the performance of the SiF_4 volatilization step. Finally, the NaOH solution, the glass funnel and the connecting polyethene tube were checked for activity with gamma spectrometric measurements. The experiments indicated that in no case had activity distilled. However, a reliable check of the glass funnel was made impossible owing to spattered Na-24 and Sc-46 activity. The experiments were therefore repeated with stock solutions of radioactive isotopes of Ag, Au, Ca, Cd, Ce, Co, Cr, Cs, Cu, Fe, Hf, La, Mo, Na, Rb, Sc, Sr, Th, U, Zn and Zr in the following, indirect way: Measured amounts of one or a few of the stock solutions were in each run mixed with 2 ml conc. H_2SO_4 and 30 mg of an inactive clay sample in a platinum dish. The mixture was then treated 6 times with 5 ml portions of HF on a hot plate as described above. After dilution with water the gamma activity of the residue was compared with that of a pipetted standard sample by means of gamma spectrometry. These experiments showed that traces of the 21 elements mentioned above were all retained to better than 98 per cent in the sulphuric acid solution.

Every waste solution resulting from different stages of the chemical separation procedure were checked for possible gamma activity. In the case of some samples the effluent from the ion exchange column constituting group 5 in fig. 1 contained Ba-131 and La-140 activity. Obviously $BaSO_4$ has been precipitated and filtered together with metallic gold on the preceding G-4 glass filter disc. The weight concentrations of lanthanum given in the tables were calculated by including also the La-140 activity in the effluent from the group 5 column, whereas quantitative determinations of Ba were omitted.

3.7. <u>Results</u>

The method was first tested on samples from bore-hole 629, representing four different depths and stages:

1.80 m fresh-water in the present lake

6.20 m salt water in the younger sea-fiord

9.00 m fresh-water in the ancient lake

9.80 m salt water in the older sea-fiord.

To get an idea of the precision of the method, four analyses were made on samples from the 1.80 m depth. As these samples were taken out and treated separately, the values obtained from this reproducibility test include not only the error of the separation method but also the errors caused by any heterogeneities of the sample as well as errors introduced in the irradiation step. (The samples were irradiated simultaneously close to each other in the same irradiation can.) The mean of the four determinations is used for the comparison with the results from the other stages. Altogether 30 elements were quantitatively determined and the results are given in table 2.

As a second step some elements, namely Br, Hg, Mn and Se, which seemed to show a difference in concentration between the marine and the fresh-water layers, were analyzed in some additional samples from borehole 629. As and Sb were also included as they could easily be analyzed simultaneously. The results are shown in fig. 3.

To verify the validity of the observations, the same elements, excluding Hg, were also analyzed in boreholes 1511 and 16 from two other sections. As in this case the irradiations had to be performed in a lower flux in the reactor R1, the activity of Hg was considered too low for a satisfactory determination. The results of these analyses are shown in figs. 5 and 6.

4. GEOLOGIC APPLICATION AND INTERPRETATION OF THE RESULTS

The results of the activation analysis are compared to the geologic development and to the properties of the various strata.

Fossils were studied to establish whether each stratum represented a sea or lake deposit (fig. 1). Among macrofossils, shells of Mollusca have been used as salinity indicators and among microfossils, especially the diatoms.

To check that the fresh-water layer of the older lake is synchronous in the three sections, pollen analysis has been used. The beginning of the Alnus graph (A_o , representing the oldest occurence of alder) represents a synchronous level in the older fresh-water layer before the transgression which left the marine layer of the younger sea-fiord. The A_o -level in the Viskan Valley has been radiocarbon dated to 6,500 B.C. [9, 1962].

In order to supplement the ocular classification of layers, their percentage compositions were determined according to a microscopic method [10, 1927]. The following charts give the percentages of fine detritus, coarse detritus, dy (dy = humus flocculations), diatom shells,

chitin and mineral particles, as well as giving the medium and maximum sizes (diameters) of the mineral grains.

We may consider the different sections and their boreholes separately.

4.1. Borehole 629

The sequence of strata (fig. 1, bottom section) shows two fiord stages and two lake stages. Except for small litoral sandbeds on the steep sides of the valley, the sediments in this narrow section of the valley have been deposited in deep water, without admixture of alluvial fill.

The strata, which according to fig. 3 are composed of muddy clay (mud, here = fine detritus gyttja), are remarkably uniform at different depths. Sample series No. 629 is therefore well suited for testing the possibility of using activation analysis to distinguish layers deposited in fresh and salt water.

The statistical investigation of the diatoms involves a determination of different species and a classification of these according to demands on the environment. Fig. 3, column 3, shows the percentages of marine diatoms at the levels where activation analysis has been made. The diatom analyses were carried out by Dr. M.B. Florin in connection with the Viskan investigation, and they have been published in a paper in which the results of the diatom analysis have been compared with spectrographic determinations of B [2, 1945].

According to table 2 only Hg and Ca have higher concentrations in the two marine samples than in the two fresh-water samples, and only Mn and U have higher concentrations in both fresh-water samples than in both marine samples. When comparing these values differences of less than four standard deviations were ignored.

The post-glacial transgression, represented by sample 6.20 m i table 2 is the best known and most interesting marine stage. If the trace-element quantities of this marine layer are compared to the quantities of the fresh-water layers below and above, the following elements also show higher concentration: Sb, As, Br and Se. On the other hand, Cs, Co, Na, Rb, W and Zn have higher concentrations in the fresh-water layers.

Thus 14 or 30 trace elements have turned out to be of interest for more intensified studies in the sections from the Viskan Valley. From methodic points of view, however, it has been convenient to test 6 elements (Hg, Mn, Sb, As, Br and Se). As seen in fig. 3 the activation analysis of Br and Se shows a fluctuation which is compatible with the salt water of the post-glacial transgression (the younger sea-fiord) and the fresh-water of the lake stages before and after the transgression. The brackish water of the older sea-fiord, however, is not registered by Br or Se. In the transition zone between the older sea-fiord and the older lake there is a distinct peak in the Mn graph.

In fig. 4 a collation has been made of the results of three different methods: salinity according to diatom analysis, boron according to spectral analysis, and bromine according to activation analysis. The results of these three methods are fairly uniform. The graph of the activation analysis gives a good picture of the course of a transgression with salinity first increasing and then decreasing.

4.2. Borehole 1511

Borehole 1511 is in a wider part of the Viskan Valley, illustrated by one of the sections of fig. 1. The section with borehole 1511, as distinguished from the previous section with borehole 629, shows an increase in the percentage and size of mineral particles from the deeper layers to the shallower ones: clayey mud (gyttja), silty mud, muddy silt and fine sand. The last mentioned are delta beds.

In borehole 1511 the record of the changes in salinity according to fig. 5 is based on various types of fossils, for the changes of salinity are generally not reflected as different strata except for the lowest marine bed. The clayey mud deeper than 9.0 m below the ground surface is characterized by shells of Mytilus edulis, which lived in the brackish water of the older sea-fiord. Between the levels 7.5 and 5.5 m below the surface appears another layer with shells of brackishmarine Mollusca such as Mytilus edulis, Cardium edule and Hydrobia ulvae. Above the layer of shells of the younger sea-fiord there are no shells to be found, though there are plenty of fossils of Nymphaea, microscopic remnents as well as seeds. These fossils indicate freshwater, probably lake water or water from the Viskan which gave rise to the sandy delta beds. Thus in borehole 1511 there are four different stages of salinity, as in borehole 629. This geologic development in 1511, based on fossil Mollusca and plants, is confirmed by fossil diatoms according to von Post's posthumous manuscript, which is to be published.

As in borehole 629 the marine layer of the younger sea-fiord has larger amounts of Br and Se than the fresh-water layers below and above the marine layer. The brackish layer of the older sea-fiord is not registered as the brackish-marine layer of the post-glacial transgression in the younger sea-fiord.

In the transition zone between the older sea-fiord and the older lake stage there is a pronounced peak in the Mn graph exactly as in borehole 629. There is, however, no Mn peak in the transition zone between the younger sea-fiord and the younger lake stage, nor in borehole 629. Thus the peaks cannot have been caused only by changes from sea to lake. When the older sea-fiord turns into the ancient lake, the black sulphurous sediment turns into a less sulphurous clay. Under the reducing circumstances in the stagnant bottom water of the older fiord, Mn was not deposited. The Mn peak appears at the very first stage of the ancient lake. In the lake, without stagnant bottom water, the more oxidizing environment favours precipitation of manganese.

4.3. Borehole 16

The section with borehole 16 is in the shallow, innermost part of the fiord, where the river Viskan has given rise to delta beds of various ages. As usual the oldest layer is the marine clay of the older sea-fiord with shells of Mytilus. Above that come two generations of delta beds separated by brown mud. The older delta bed contains fossil pollen of the fresh-water plant Myriophyllum alterniflorum living in the ancient lake. On the delta plains arose small basins which were filled with deposits of fen peat with remnants of Phragmites and brown mud rich in humus (dy). This mainly organic layer was later covered by the sea during the post-glacial transgression. The younger delta bed of borehole 16 was deposited in the brackish water of the younger sea-fiord, made sweet by the river water.

The two delta beds over the bottom marine bed show fairly similar quantities of trace elements. Br and Se have somewhat higher values in the upper part of the brown mud characterized by coarse detritus such as leaves and sticks.

5. STATISTICAL TREATMENT

To get an idea of the influence of grain size on trace element content of the sediments, a division is made between argillaceous and sandy sediments in the table below. The investigated sandy sediments were deposited in fresh-water as fluvial deltas. In the investigated sandy samples more than half of the volume consists of mineral particles and the mean size is larger than 20 μ .

Collocation of sandy and argillaceous sediments deposited in fresh-water:

	Bore- hole	-				
		As ppm	Br ppm	Mn %	${ m \tilde{Sb}}$ ppm	Se ppm
Sandy sediments:						
present lake	1511	4.3	7.4	0.05	0.28	0.18
ancient lake	16	3.9	4.4	-	0.14	0.24
younger sea-fiord (fresh-water)	16	3.4	10.2	-	0.14	0.23
Argillaceous sediments	5:					
present lake	629	5.9	5.3	0.14	0.51	0.65
ancient lake	629	6.4	15.2	0.18	0.43	0.67
- ¹¹ -	1511	7.7	20.3	0.17	0.49	0.53

As we do not have the same boreholes in the two groups of the table, direct comparisons are impossible. However, the means give us reason to suspect that there may be higher concentrations of the in-

vestigated elements in argillaceous than in sandy sediments. If the same also applies to marine layers, argillaceous and sandy layers are not comparable for discrimination of the salinity. For this reason the sandy delta layers have been excluded from the following statistical treatment. All other values have been grouped according to environment (Table 3).

The fresh-water sediments are divided into present and ancient lake. The samples from the fen and from the mud in borehole 16 are separated from argillaceous samples.

The salt-water sediments are grouped according to different salinity. The samples from the younger sea-fiord must be divided into different groups. In borehole 16 the fiord has been made sweet by river water. Borehole 1511 is more brackish than 629, which is situated farther out in the younger sea-fiord. As the post-glacial transgression implied increased depth and a larger amount of salt and as the subsequent regression led to reversed conditions, layers of different salinity were deposited in the younger sea-fiord, here divided into three groups: brackish, brackish-marine and marine. The samples from the older sea-fiord have been referred to the brackish samples.

Analysis of variance (one-way layout with fixed effects) was used for statistical investigation of all values except those from the sandy layers. If the hypothesis has been rejected (F-test) that all cell means are equal (H_0), the Scheffé-method [11] has been applied for judging all contrasts to find out if relevant contrasts are significantly different from zero at the 95 % level of significance. Relevance is here understood as a comparison of layers of different salinity in the same borehole (or boreholes). As there are rather few observations, only large differences in cell means can be discovered.

Results of the tests

 \underline{Br}

H_o is rejected (p < 0.001).

Estimated contrasts significantly different from zero:

$\bar{X}_4 - \bar{X}_5 = 50.5$	$\bar{x}_{12} - \bar{x}_6 = 96.0$
$\vec{X}_{12} - \vec{X}_1 = 109.4$	$\vec{x}_{12} - \vec{x}_8 = 95.2$
$\vec{X}_{12} - \vec{X}_2 = 99.5$	$\vec{X}_{12} - \vec{X}_{10} = 58.7$

- <u>As</u> H_0 is rejected (p < 0.001). No contrasts of relevant interest are significantly different from zero.
- \underline{Sb} H_o is rejected (p < 0.001). Estimated contrasts significantly different from zero.

$$\frac{\overline{X}_{10} + \overline{X}_{11}}{2} - \frac{\overline{X}_6 + \overline{X}_7}{2} = 0.34$$
$$\frac{\overline{X}_{10} + \overline{X}_{11}}{2} - \frac{\overline{X}_2 + \overline{X}_3}{2} = 0.36$$

Se
$$H_0$$
 is rejected (p < 0.001).

Estimated contrasts significantly different from zerc.

 $\overline{x}_{12} - \overline{x}_1 = 0.90 \qquad \overline{x}_{11} - \overline{x}_7 = 0.93$ $\overline{x}_{12} - \overline{x}_2 = 0.88 \qquad \overline{x}_{11} - \overline{x}_9 = 1.23$ $\overline{x}_{12} - \overline{x}_8 = 1.26$

<u>Hg</u> H is rejected (p < 0.05).

No relevant contrasts differ significantly from zero.

6. CONCLUSIONS

From the statistical treatment above the following hypotheses may be stated.

In the most marine layer of the post-glacial transgression <u>Br</u> has significantly higher values than in all other layers. Probably Br is also higher in the brackish marine layers compared to laustrine and brackish layers. <u>As</u> does not show any sure trend with increasing salinity. <u>Sb</u> is significantly higher in the brackish marine layer than in the brackish layer of the younger sea-fiord and the fresh-water layer of the ancient lake. <u>Se</u> is higher in the marine layer than in the layer of the older sea-fiord and the fresh-water layers. According to borehole 1511 there is also a difference between brackish-marine and brackish layers. Probably both the marine and the brackish-marine layers have higher amounts of Se than all other layers. Concerning <u>Hg</u> no significant differences have been stated. If there really are differences we suspect that the lacustrine layers have lower concentrations than the other layers.

Disregarding the special and complicated sequence of strata in borehole 16, Br and Se are of interest as saline indicators in argillaceous layers with organic detritus. In the investigated sections of the Viskan Valley Br and Se have been enriched in the younger sea-fiord layers. But concerning the older sea-fiord layers with ferrous sulphide indicating stagnant water, the analysed samples are few and the results contradictory. Possibly Br and Se were initially concentrated and later released by processes in the bottom below stagnant water.

For comparisons of trace-element concentrations in rocks, soils, animals, plants and sea-water reference is made to Bowen's new collocation and literature cited by him [12].

According to this publication Br can be very much enriched in marine plants and animals. This is interesting, as remnants of organisms constitute the organic fine detritus of the argillaceous sediments.

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Element determined	Nuclide used for the deter- mination	т 1/2	Gamma energies used for identifi- cation					
	ci 100							
Antimony	Sb-122	2.8 d	0.564					
Arsenic	As-76	26.5 h	0.55					
Barium	Ba - 131	11.6 d	0.21, 0.50					
Bromine	Br-82	36 h	0.78					
Calcium	Ca - 47	4.7 d	1.31					
Cerium	Ce - 141	33.1 d	0.145					
Cesium	Cs-134	2.4 y	0.605					
Chromium	Cr-51	28 d	0.32					
Cobalt	Co-60	5.3 y	1.33					
Copper	Cu-64	12.8 h	0.51					
Europium	Eu -1 52	13 y	1.42					
Gallium	Ga - 72	14.2 h	0.83, 2.50					
Gold	Au -1 98	2.7 d	0.41					
Hafnium	Hf -1 81	46 d	0.482					
Iron	Fe-59	45 d	1.29					
Lanthanum	La -140	40.2 h	1.60					
Lutetium	Lu-177	6.75 d	0.208					
Manganese	Mn - 56	2.6 h	0.84					
Mercury	Hg-203	45.8 d	0.279					
Phosphorus	P-32	14. 3 d	Bremsstrahlung					
\mathbf{P} latinum	Au - 199	3 .1 4 d	0.16					
Rubidium	Rb - 86	18.6 d	1.08					
Samarium	Sm-153	47 h	0.102					
Scandium	Sc-46	85 d	1.12					
Selenium	Se - 75	127 d	0.265					
Silver	Ag-110	270 d	0.66					
Sodium	Na-24	15 h	2.76					
Strontium	Sr- 85	65 d	0.51					
Tantalum	Ta ~1 82	112 d	1.12 - 1.23					
Terbium	Tb-160	73 d	0.299					
Thorium	Pa-233	27 d	0.31					
Tin	Sn-113	119 d						
	(In-113 m)	104 m	0.39					
Tungsten	W-187	24 h	0.686					
Uranium	Np-239	2.3 d	0.23, 0.28					
Ytterbium	Yb -17 5	4 .1 d	0.396					
Zinc	Zn-65	245 d	1.11					
 Zirconium	Zr-95	63.3 d	0.72, 0.75					

Table 2. Borehole 629

Element	1	Depth in meter												
determined		1.80 Present mean	t lake	6.20 Younger sea-fiord	9.00 Ancient lake	9.80 Older sea-fiord								
Antimony ppm	0.47 0.52 0.48 0.50	0.49	<u>+</u> 0.02	0.63	0.41	0.48								
Arsenic "	4.1 4.2 4.2 4.8	4.3	<u>+</u> 0.32	8.2	5.8	5.5								
Bromine "	5.9 6.7 6.3 7.7	6.7	<u>+</u> 0.77	131	18	21								
Calcium %	0.72 1.06 1.10 0.99	0.97	<u>+</u> 0.17	2.1	1.2	2.8								
Ce ri um ppm	230 250 284 207	243	<u>+</u> 32	207	192	210								
Cesium "	6.6 7.3 6.3 7.0	6.8	<u>+</u> 0.44	4.7	7.0	7.3								
Chromium "	48 58 51 54	53	<u>+</u> 4.3	47	57	70								
Cobalt "	21 22 22 20	21	<u>+</u> 1.0	11	20	20								
Europium "	2.4 3.0 2.5 2.2	2.5	<u>+</u> 0.34	2.2	3.3	2.7								
Gallium "	14 17 13 17	15	<u>+</u> 2.1	17	13	18								

Table 2. Continuation

Element		Depth in meter											
determined]	1.80 Present mean	lake	6.20 Younger sea-fiord	9.00 Ancient lake	9.80 Older sea-fiord							
Hafnium ppm	5.4 5.7 6.3 6.6	6.0	<u>+</u> 0.55	4.7	6.4	6.6							
Iron %	5.8 7.1 6.0 6.1	6.2	<u>+</u> 0.58	5.8	7.1	6.5							
Lanthanum ppm	57 59 53 53	56	<u>+</u> 3.2	55	70	60							
Lutecium "	0.89 1.08 1.03 0.80	0.95	<u>+</u> 0.13	0.6	1.3	1.3							
Manganese %	0.130 0.127 0.128 0.129	0.128	<u>+</u> 0.0014	0.058	0.109	0.059							
Mercury ppm	0.36 0.36 0.39 0.33	0.36	<u>+</u> 0.025	2.0	0.81	1.4							
Rubidium "	109 117 110 115	113	<u>+</u> 3.9	90	129	145							
Samarium	15 18 13 14	15	<u>+</u> 2.2	15	20	13							
Scandium "	14 17 14 15	15	<u>+</u> 1.4	10	13	15							
Selenium "	0.68 0.78 0.75 0.67	0.72	<u>+</u> 0.053	2.0	1.0	0.32							

<u>Table 2.</u> Continuation

Element	Depth in meter											
determine	:d	I	1.80 Present mear	lake	6.20 Younger sea-fiord	9.00 Ancient lake	9.80 Older sea-fior					
Sodium	70	1.11 1.17 1.16 1.16	1.15	<u>+</u> 0.027	0.96	1.43	1.39					
Strontium]	ppm	81 116 131 132	115	<u>+</u> 24	160	191	217					
Tantalum	t t	1.16 1.43 1.04 1.25	1.2	<u>+</u> 0.17	1.2	1.0	-					
Terbium	11	0.84 1.13 0.88 0.93	0.95	<u>+</u> 0.13	1.1	1.5	1.6					
Thorium	t t	14 14 16 13	14	<u>+</u> 1.3	12	15	15					
Tungsten	JJ	1.5 1.6 1.5 1.5	1.5	<u>+</u> 0.058	1.2	1.6	1.6					
Uranium	ן נו	6.1 6.3 6.3 6.5	6.3	<u>+</u> 0.16	4.8	11.2	4.7					
Ytterbium	31	1.9 2.8 2.4 2.9	2.5	<u>+</u> 0.45	1.9	2.9	3.1					
Zinc	t1	116 122 120 112	118	<u>+</u> 4.5	87	143	117					
Zirconium	†1	295 349 264 327	309	<u>+</u> 37	253	423	230					

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Table 3

	Environment	Bore-			As			Br			· · · · ·	Hg]		Mn				Sb				Se	
1		hole	ⁿ i	¥ (ppm)		^s i ²	ⁿ i	X ₁ (ppm)	s1 ²	ⁿ i	ī, (ppm)		^s 1 ²	ⁿ i	₹ <u>1</u> (%)		*1 ²	ⁿ i	X _i (ppm)		5 ₁ 2	n _i	x _i (ppm)		8 ₁ 2
	Lacustrine:																								
1.	present lake	629	3	5.9	<u>+</u> 2.7	7.21	3	5.3 <u>+</u> 1.8	3.24	3	0.33	<u>+</u> 0.10	0.009	3	0.137	<u>+</u> 0.012	0.00014	3	0.51	<u>+</u> 0.06	0.0033	3	0.65	<u>+</u> 0.03	0.0010
2.	ancient lake	629	4	6.4	<u>+</u> 1.7	3.02	5	15.2 <u>+</u> 4.3	18.36	4	2.19	<u>+</u> 2.83	7.92	5	0.182	<u>+</u> 0.145	0.0210	4	0.43	<u>+</u> 0.06	0.0035	4	0.67	<u>+</u> 0.18	0.0323
3.	-"-	1511	3	7.7	<u>+</u> 2.7	7.39	3	20.3 <u>+</u> 3.8	14.34	-	-	-	-	3	0.170	<u>+</u> 0.182	0.0331	3	0.49	<u>+</u> 0.08	0.0057	3	0.53	<u>+</u> 0.10	0.0102
4.	fen	16	3	2.4	<u>+</u> 0.5	0.28	3	58.7 <u>+</u> 35.8	1284.3	-	-	-	-	-	-	-	-	3	0.14	<u>+</u> 0.04	0.0012	3	0.65	<u>+</u> 0.34	0.1159
5.	mudi	16	3	10.3	<u>+</u> 1.5	2.29	3	8.2 <u>+</u> 2.0	3.91	-	-	-	-	-	-	-	-	3	0.28	<u>+</u> 0.02	0.0006	3	0.23	<u>+</u> 0.02	0.0003
	Brackish																								
6.	transition lake	629	2	14.7	<u>+</u> 0.3	0.12	2	18.7 <u>+</u> 3.7	14.04	2	7.90	<u>+</u> 4.40	19.28	2	0.150	<u>+</u> 0.042	0.0018	2	0.44	<u>+</u> 0.26	0.0684	2	0.73	<u>+</u> 0.14	0.0200
7.	_"_	1511	4	9.2	<u>+</u> 1.6	2.59	4	18.3 <u>+</u> 3.6	12.92	-	-	-	-	4	0.060	<u>+</u> 0.045	0.0020	4	0.51	+0.01	0.0091	4	0.47	<u>+</u> 0.09	0.0078
8.	older sea-fiord	629	1	5.5	-	-	2	19.5 <u>+</u> 2.6	6.48	1	1.36	-	-	2	0.055	<u>+</u> 0.007	0,00005	1	0.48	-	-	1	0.29	-	-
9.	-"-	1511	4	11.6	±3.2	10.32	4	13.0 <u>+</u> 2.5	6.00	-	-	-	-	4	0.123	<u>+</u> 0.075	0.0056	4	0.65	<u>+</u> 0.05	0.0022	4	0.17	<u>+</u> 0.25	0.0625
	Brackish-marine																					1			
10.	younger sea-fiord	629	2	9.8	-	-	2	46.0 <u>+</u> 9.9	99.4	2	7.75	<u>+</u> 0.64	0.41	2	0.070	<u>+</u> 0,014	0.0002	2	0.90	<u>+</u> 0.12	0.0144	2	0.95	<u>+</u> 0.25	0.0612
11.	-"-	1511	2	14.5	<u>+</u> 0.5	0.30	2	47.0 ± 5.7	32. 0	-	-	-	-	2	0.065	<u>+</u> 0.007	0.00005	5	0.73	<u>+</u> 0.04	0.0018	2	1.40	<u>+</u> 0.14	0.0200
	Marine																								
12.	younger sea-fiord	629	4	9.5	±3.2	10.17	4	114.7 <u>+</u> 16.2	262.1	4	5.25	±3.17	10.06	4	0.083	<u>+</u> 0,021	0.00042	4	0,61	<u>+</u> 0.15	0.0215	4	1.55	<u>+</u> 0.26	0.0668

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n_i = number of observations

 \bar{X}_{i} = arithmetic mean

 s_1^2 = estimated variance





Fig. 2



BORE HOLE 629

v. *** 4







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Fig. 6.



DIAGRAM I





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CLAY SAMPLE GROUP 4: SULPHIDE PRECIPITATE COOLING PERIOD : 10 DAYS COUNTING TIME : 40 MIN



























CLAY SAMPLE CROUP 20: DOWEX 2-50 MIXED BED COOLING PERIOD: 4 DAYS COUNTING TIME: 20 MIN



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