

# U, Th and K in Upper Cretaceous and Tertiary sediments in Denmark

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Engell-Jensen, M., Korsbech, U. & Madsen, F. E.: U, Th and K in Upper Cretaceous and Tertiary sediments in Denmark. *Bull. geol. Soc. Denmark*, vol. 32, pp. 107-120. Copenhagen, September, 4th, 1984. <https://doi.org/10.37570/bgscd-1983-32-07>

The paper presents the results of the measurements of the equivalent equilibrium concentrations of the radioactive elements uranium, thorium, and potassium in about 500 samples of Danish sedimentary rocks from Campanian to Miocene. The results are presented as cross-plots in which each sample has been marked according to its pairwise contents of the radioactive elements. The selected presentation of the results may form a basis of a lithostratigraphical interpretation of the SNG-log (Spectral Natural Gamma-ray log) which is a borehole measurement of the equivalent equilibrium concentration of U, Th, and K in the present formations. Some types of sediments contain distinct concentrations of uranium, thorium, and potassium and could easily be identified from a SNG-log. For other sediments the identification cannot be based on the cross-plots alone, but other information has to be included. A SNG-log from a borehole penetrating 55 meters of Danian calcarenite is presented as an example of the use of the SNG-log for stratigraphical subdivision and correlation in sediments where other methods would be difficult and time consuming to perform.

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## Introduction

In connection with deep drilling, wireline logging has for a long time played an essential part in the lithological subdivision and the evaluation of the penetrated formations, and for shallow borings the logging methods are becoming important too.

Open-hole well logging includes electrical, acoustical, and nuclear methods. In cased holes only nuclear methods based on neutrons and gamma-rays can be used. The latter are consequently the logging methods applied in old wells or in shallow wells, where casing is mandatory due to the un lithified formations.

The natural gamma-ray log is one of the most common of the wireline logs. It is a recording of the total intensity of gamma-rays penetrating the borehole from the formation. Gamma-rays are emitted during the radioactive decay of potassium-40 together with uranium-238 and thorium-232 and their radioactive daughters.

Within the latest ten years the development of better nuclear detectors, electronics, and micro computers has formed the basis of the spectral natural gamma-ray log (SNG-log). With a SNG-

probe it is possible to separate the gamma-rays in a borehole into three parts. One part is the gamma-rays coming from uranium and daughters, the other part is the gamma-rays from thorium and daughters, and the third part is the gamma-rays from potassium.

A SNG-log measured with a calibrated SNG-probe is presented as curves showing the concentration of Th, U, and K in the surrounding formation.

The lithostratigraphical interpretation of the SNG-log requires a knowledge of the concentrations of K, U, and Th in different sedimentary formations.

Investigations on this subject have been carried out in the laboratory by – among others – Adams & Weaver (1958), Hassan, Hossin & Combaz (1976), and Berstad & Dypvik (1982).

Hassan et al. found that in clean carbonate rocks U is the only radioactive element present. In marl both U and small amounts of K and Th were found. Enrichment of U in organic matter and phosphate was demonstrated by the fission track technique. Th was found to be mainly associated with the clay minerals. However, in

sandstones heavy minerals in sufficient quantities are manifested by an excessive Th content.

Berstad & Dypvik studied a Tertiary sedimentary sequence of a well in the central North Sea. They found that Th and K were associated with the clay fraction, while U was enriched in the silt fraction. In the volcano-clastic Paleocene and Eocene beds the low gamma-activity was due to the low content of U and Th in the volcanic debris.

The aim of the present work has been to find – if possible – the relations between the lithology and the distribution of the radioactive elements in a number of Danish sedimentary formations.

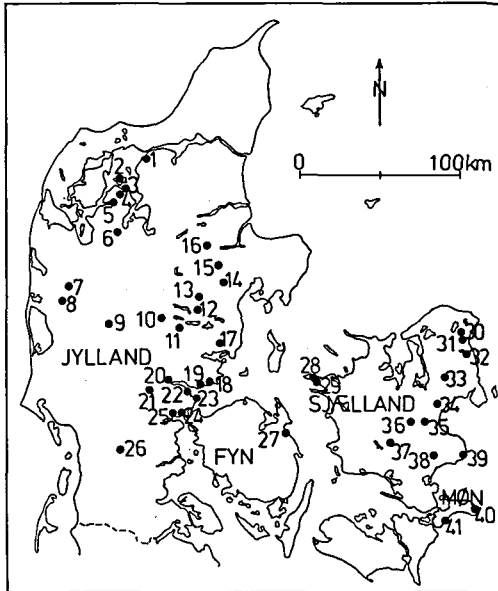


Fig. 1. The dots indicate the sampling localities for the study. The localities are:

1. Aggersund, chalk quarry. -2. Fuur, cliffs and Mo-clay pits. -3. Branden, clay pit. -4. Batum, chalk quarry. -5. Harre, borehole. -6. Skive, clay pit. -7. Lille Spåbæk, clay pit. -8. Muldbjerg, drilling. -9. Søby, lignite pit. -10. Salten profile. -11. Addit, gravel pits. -12. Nr. Vissing, clay pit. -13. Låsby, gravel pit. -14. Grundfør, clay pit. -15. Ølst, clay pit. -16. Svejstrup, marl pit. -17. Ås, marl pit. -18. Juelsminde, cliffs. -19. Albæk Hoved, cliff. -20. Greisdalen, gravel pit. -21. Vork, borehole. -22. Brejning, cliffs. -23. Trelde Næs, cliff. -24. Børup, cliff. -25. Hagenør, cliff. -26. Gram, clay pit and borehole. -27. Lundsgård Klint, cliff. -28. Slettenshage, clay pit. -29. Røsnæs, cliffs. -30. Langstrup, borehole. -31. Nivå, borehole. -32. Nebbegård, boreholes. -33. Hove, chalk quarry. -34. Karlstrup, chalk quarry. -35. Lellinge, river. -36. Regne-mark, borehole. -37. Glumsø, surface exposure. -38. Fakse, chalk quarry. -39. Stevns Klint, cliff. -40. Møns Klint, cliff. -41. Hvideklint, cliff.

	FORMATION	
MIOCENE	GRAM FM	
	HODDE FM	
	ARNUM FM	ODDERUP FM
	KLINTINGHOVED FM	RIBE FM
		VEJLE FJORD FM
OLIGOCENE	BRANDEN FM	
	VIBORG FM	
EOCENE	SØVIND MARL FM	
	LILLEBÆLT CLAY FM	
	RØSNES CLAY FM	
	ØLST FM	FUR FM
PALEOCENE	KERTEMINDE MARL FM	
	LELLINGE GRS FM	
DANIAN	LIMESTONE	
MAASTRICHT AND CAMPANIAN	WHITE CHALK	

Fig. 2. Lithostratigraphical profile of Danish formations from Campanian to Miocene.

The basis of the work has been a total of 500 samples collected from surface exposures and shallow borings. The samples have been characterized with respect to their lithology and their content of radioactivity has been measured.

The method of sampling has limited the investigations to the geological periods from Upper Campanian to Upper Miocene.

## Sampling and measurements

Fig. 1 shows the locations from where the samples have been collected, and fig. 2 shows a lithostratigraphical profile of all major Danish formations from Upper Campanian to Upper Miocene. We have tried to include in the study samples from all formations. Only samples from the Ribe Formation and the Arnum Formation have not been measured.

Geological surveys of the abovementioned formations are found in Dinesen, Michelsen & Lieberkind (1977), Radwanski, Friis & Larsen (1975), Dinesen (1976), Thiede, Nielsen &

Perch-Nielsen (1980), and Christensen & Uileberg (1973).

The gamma-activity of the samples has been measured with a Ge(Li) gamma-detector. Both U with daughters and Th with daughters emit gamma-radiation of several energies, and the measurements of both U and Th have been based on gamma-radiation of four different energies.

The Th measurements have been based on the gamma-radiation emitted from actinium-228 and thallium-208, which are radioactive daughters of thorium-232. The U measurements have been based on the gamma-radiation emitted from uranium-235, radium-226, lead-214, and bismuth-214. In general Th and U are in radioactive equilibrium with their daughters. For U, however, the equilibrium may have been strongly disturbed through geochemical processes because of the long life-times of some of the radioactive daughters. In those cases the true concentration of uranium differs from the measured equivalent equilibrium concentration of uranium, i.e. that concentration of uranium in equilibrium with its daughters that has the same spectral gamma-radioactivity as the actual sample. (Equivalent equilibrium concentration of uranium is also denoted gamma equivalent uranium or simply equivalent uranium).

A SNG-probe also measures the equivalent equilibrium uranium concentration because a SNG-measurement of uranium in general is based on the measurement of bismuth-214.

"Equivalent equilibrium uranium concentration" is in the following abbreviated to eU concentration and for thorium the abbreviation is eTh.

Among the potassium isotopes only potassium-40 emits gamma-radiation, which can be measured in the laboratory and in a borehole.

In general Danish sedimentary rocks contain low concentrations of radioactive elements. In order to obtain acceptable counting statistics, most samples have been measured for at least 6 hours. Samples with very low concentrations of radioactive elements, e.g. Danian Limestone, have been measured for 24 hours.

The volume of each sample was 850 ml.

The accuracy of the measurements has in general been better than 3% for K, 5% for eTh, and 10% for eU. All concentrations are measured relative to dry samples (16 hours at 105°C).

## Cross-plots

The results of the measurements are presented as crossplots in the figures 3 to 8. The figures 3 to 5 concern older Tertiary sediments and White Chalk of Cretaceous age, whereas the figures 6 to 8 concern younger Tertiary sediments. In a cross-plot each sample is assigned a position according to its concentration of K, eU, and/or eTh. When all samples of a certain type have been plotted, one usually finds that they are confined to a limited area within the figure. When all samples of all types have been plotted, some types occupy their "own" area of the cross-plot, whereas other types of samples intermix with each other.

In order to simplify the "readings" of the cross-plots, the areas corresponding to each type of samples – i.e. each type of sediment – have been marked. These marked areas should not be taken too rigorously. A few atypical samples could be found outside the marked areas. Further it is possible that new samples taken from a new locality may not fit exactly within the marked areas determined through this study.

In the first cross-plot, fig. 3 most of the samples fall into three main areas: carbonates, sticky and plastic clay, and a diatomite called Mo-clay. (In this paper sticky and plastic clay is equivalent to Eocene clays – see fig. 2).

The carbonates – White Chalk, Danian Limestone and Calcarene – contain minor amounts of silicious material. The calcareous Lelling Green sand further contains glauconite, which accounts for a major fraction of the K content.

In the sticky and plastic clay illite is present and contribute to the high K content (Tank 1963). The Mo-clay from the Fur Formation has been deposited in a dominantly reducing environment together with organic material and this is reflected in the high concentration of eU (Bonde 1973; Pedersen 1981).

In fig. 4, which is a Th-K cross-plot, one finds – as expected – low concentrations of eTh in the carbonates. The higher concentrations of eTh and K in the Kerteminde Marl are due to the content of clay. Sticky and plastic clay and the Mo-clay both show high concentrations of eTh. In the sticky and plastic clay the high concentration of eTh reflects the high clay mineral content. In the Mo-clay the clay mineral fraction is in average lower, and the high concentrations

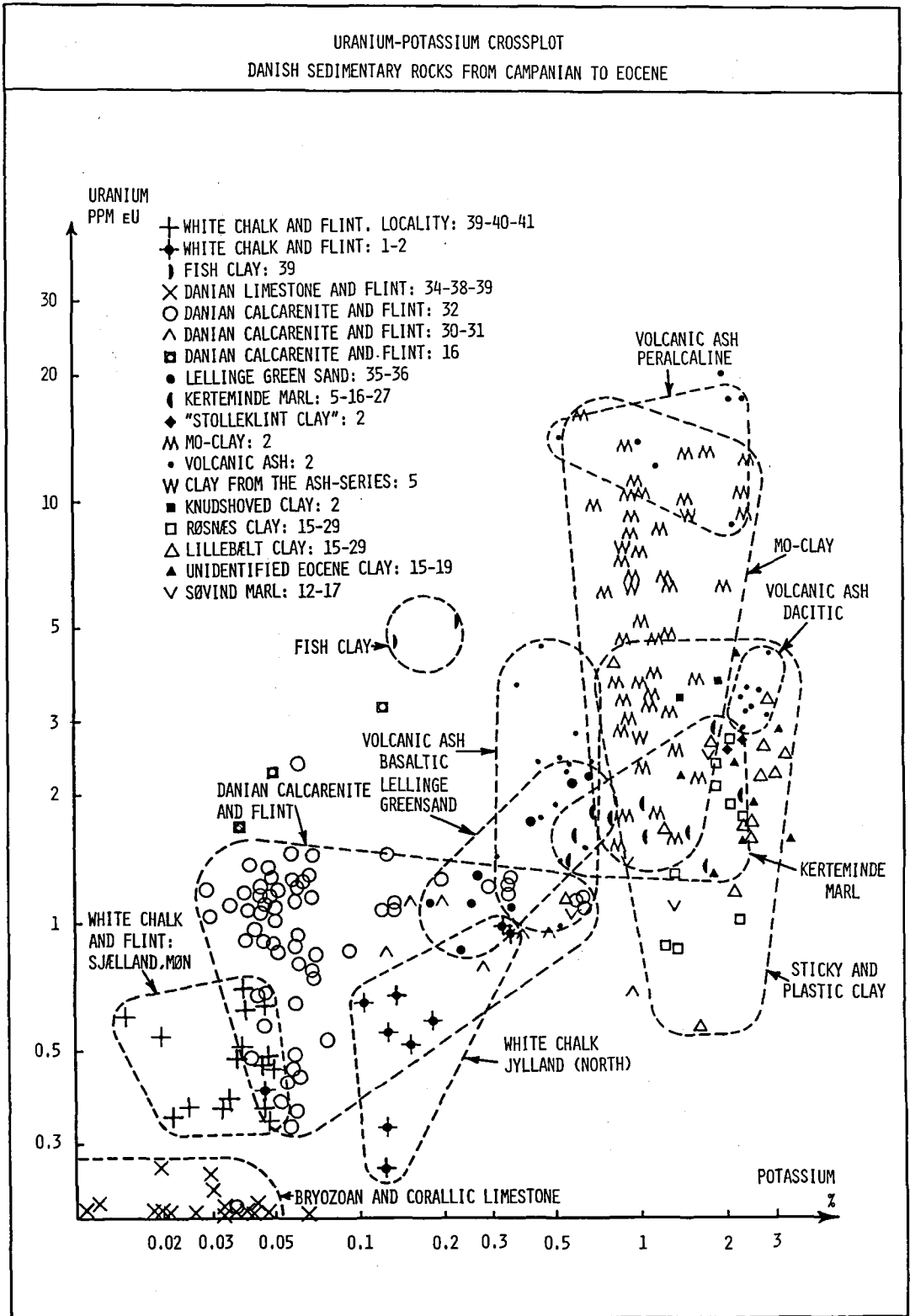


Fig. 3. Uranium-potassium cross-plot for Cretaceous White Chalk and Danian, Paleocene and Eocene sediments.

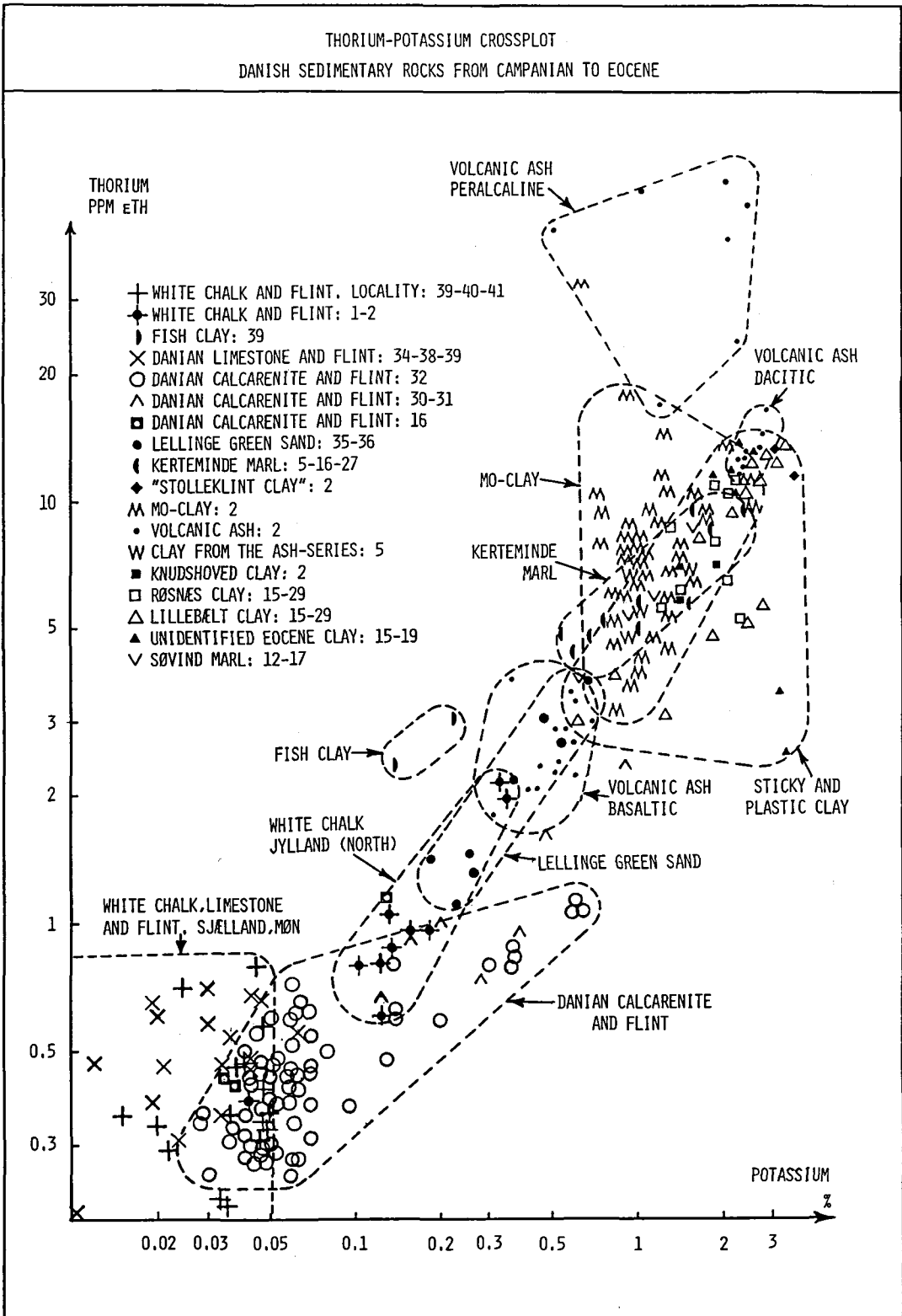


Fig. 4. Thorium-potassium cross-plot for Cretaceous White Chalk and Danian, Paleocene and Eocene sediments.

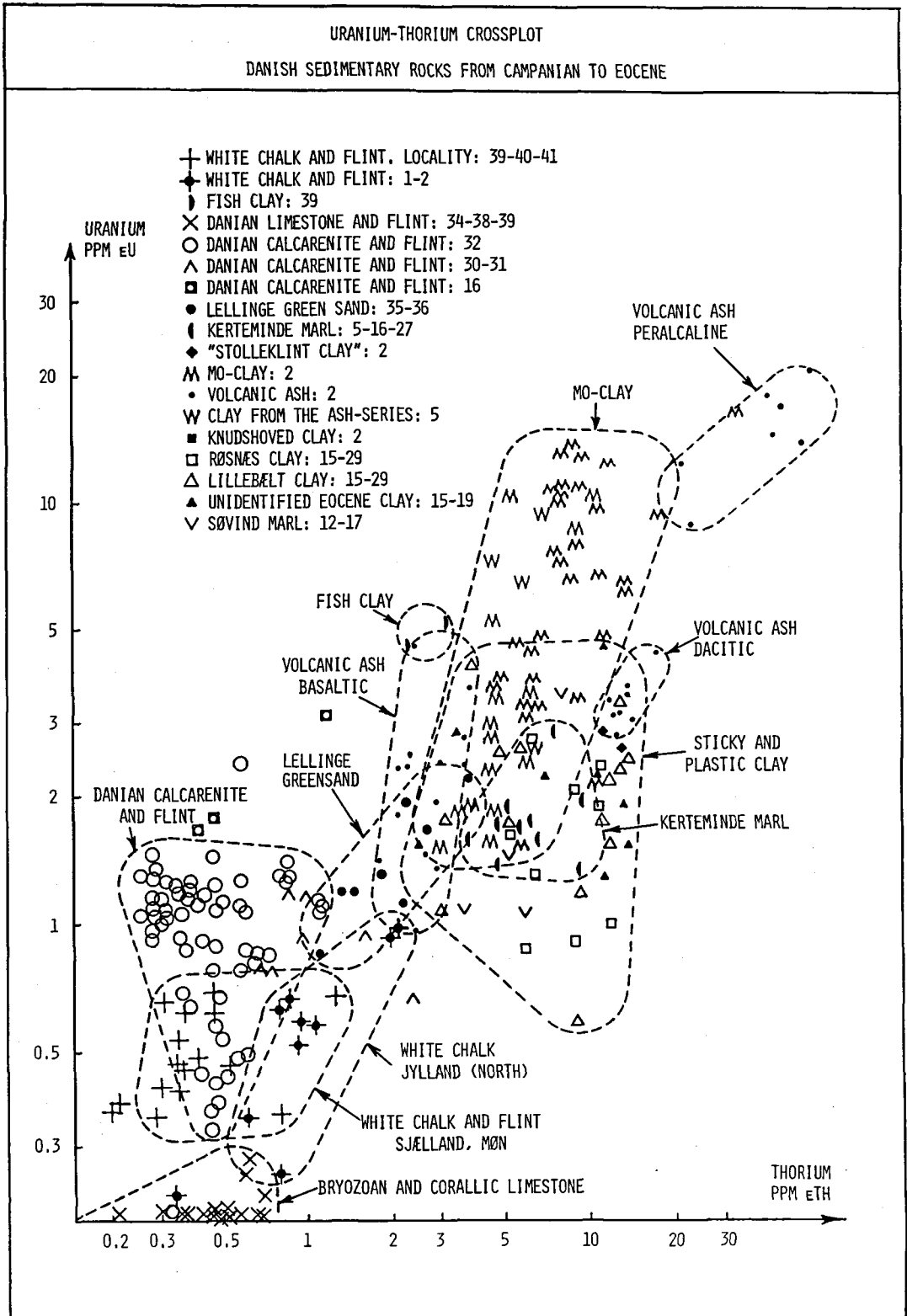


Fig. 5. Uranium-thorium cross-plot for Cretaceous White Chalk and Danian, Paleocene and Eocene sediments.

of eTh has not yet found a satisfactory explanation. Perhaps it is related to the high volcanic activity during the Late Paleocene and the Early Eocene. Especially the peralcaline and dacitic ash layers in the oldest part of the ash series contain high amounts of eTh. Volcanic debris originally deposited outside the Mo-clay area could later have been transported to the Mo-clay area and deposited together with the diatoms.

The U-Th cross-plot of fig. 5 shows a distribution pattern in accordance with fig. 3 and fig. 4.

The Danian Limestone of coralline or bryozoan origin contains the lowest concentrations of both eTh, eU, and K. The very low concentrations of eTh and K indicate that the supply of clay particles has been very low compared to the rate of accumulation of the calcareous components during the deposition of the Limestone. The low concentration of eU indicates an oxidizing environment during the sedimentation or possibly later.

The samples of flint taken from the Danian Bryozoan Limestone also have very low concentrations of radioactive elements.

The White Chalk and flint are of Campanian and Maastrichtian age. The samples collected from Sjælland and Møn differ from the samples from the northern and eastern part of Jylland with respect to the concentrations of radioactive elements. The samples from Jylland have higher concentrations of K and eTh than the samples from Sjælland and Møn. The concentrations of eU do not differ much. This indicates a higher percentage of clay particles in the White Chalk from Jylland. The few samples of Campanian age (Møn) do not differ from the samples of Maastrichtian age from the same area.

A few samples of coccolitic Danian Chalk from the North Sea are similar to the samples of White Chalk from Jylland. (The samples from the eastern part of Jylland and the samples from the North Sea are not shown in the cross-plots).

Samples of flint from the White Chalk have concentrations of the radioactive elements similar to that of the surrounding chalk.

The samples of Danian Calcarenite originates from a few water-supply borings in the northern part of Sjælland; most of the samples are from Nebbegård just north of Copenhagen.

As is seen from the cross-plots, the concentrations of K, eU, and eTh are low and vary

extensively. SNG-logs from three borings at Nebbegård provide an explanation hereof (see fig. 9). From 35 m to 90 m below the surface thick layers of rather homogeneous calcarenite and flint are separated from each other by thin marker horizons with higher concentrations of radioactivity – especially eU.

The samples included both calcarenite and flint, and here too the flint contains roughly the same amounts of radioactive elements as the calcarenite. Higher concentrations of eU have, however, not been found in the flint.

20 samples – not indicated in the cross-plots – of lower Danian calcarenite from the eastern part of Jylland (Mariager) have concentrations similar to the samples from Sjælland.

The Fish Clay from Stevns (Sjælland), which is a thin stratum separating the Cretaceous from the Tertiary, contains higher concentrations of eTh and eU and a comparatively low concentration of K. The Lellinge Greensand is from Middle Paleocene and consists of carbonates, glauconite, and a minor amount of quartz grains. The glauconite contains potassium and the uranium may be associated with the carbonates or the glauconite. The content of thorium has probably been brought to the area together with the precursors of the glauconite.

Kerteminde Marl from Middle Paleocene could be considered a mixture of clay and carbonates – and has concentrations of the radioactive elements in accordance with this.

The area “Sticky and Plastic Clay” in the cross-plots includes several types of clay. The concentrations of eTh, eU, and K seem to vary in a non-systematic way, and it has not been possible to assign specific areas of the cross-plot to different types of clay.

The Mo-clay – a marine diatomite – is found only in the north-western part of Jylland. It has been deposited in the Late Paleocene and/or the Early Eocene and it is contemporary with the Volcanic Ash also shown in the cross-plots. The concentrations of eU and eTh in the Mo-clay vary extensively.

It is generally assumed that the Mo-clay has been deposited in a dominantly non-oxidizing environment with some shorter more oxid periods (Bonde 1973; Pedersen 1981). This is in accordance with the high – and varying – concentrations of eU.

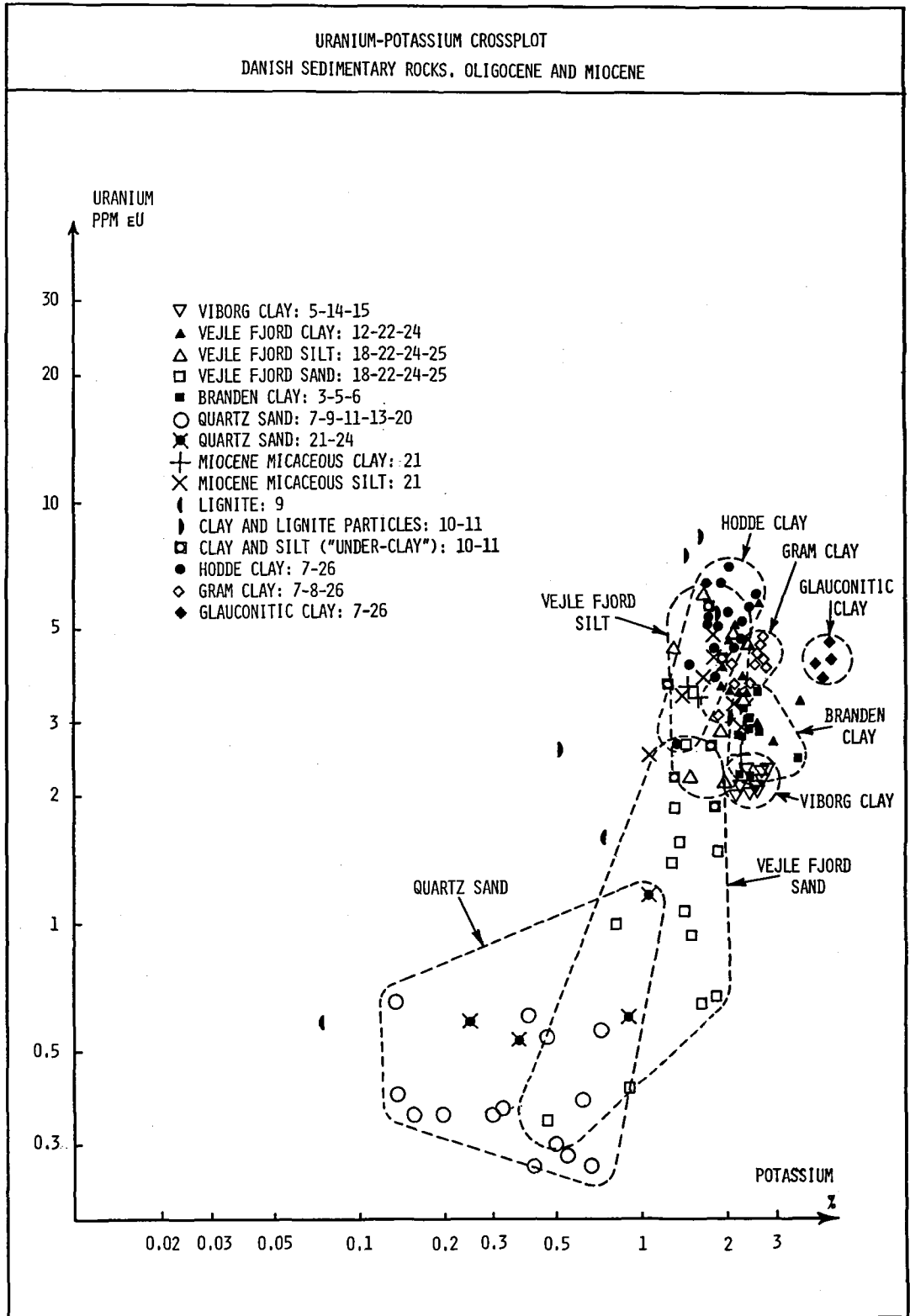


Fig. 6. Uranium-potassium cross-plot for Oligocene and Miocene sediments.



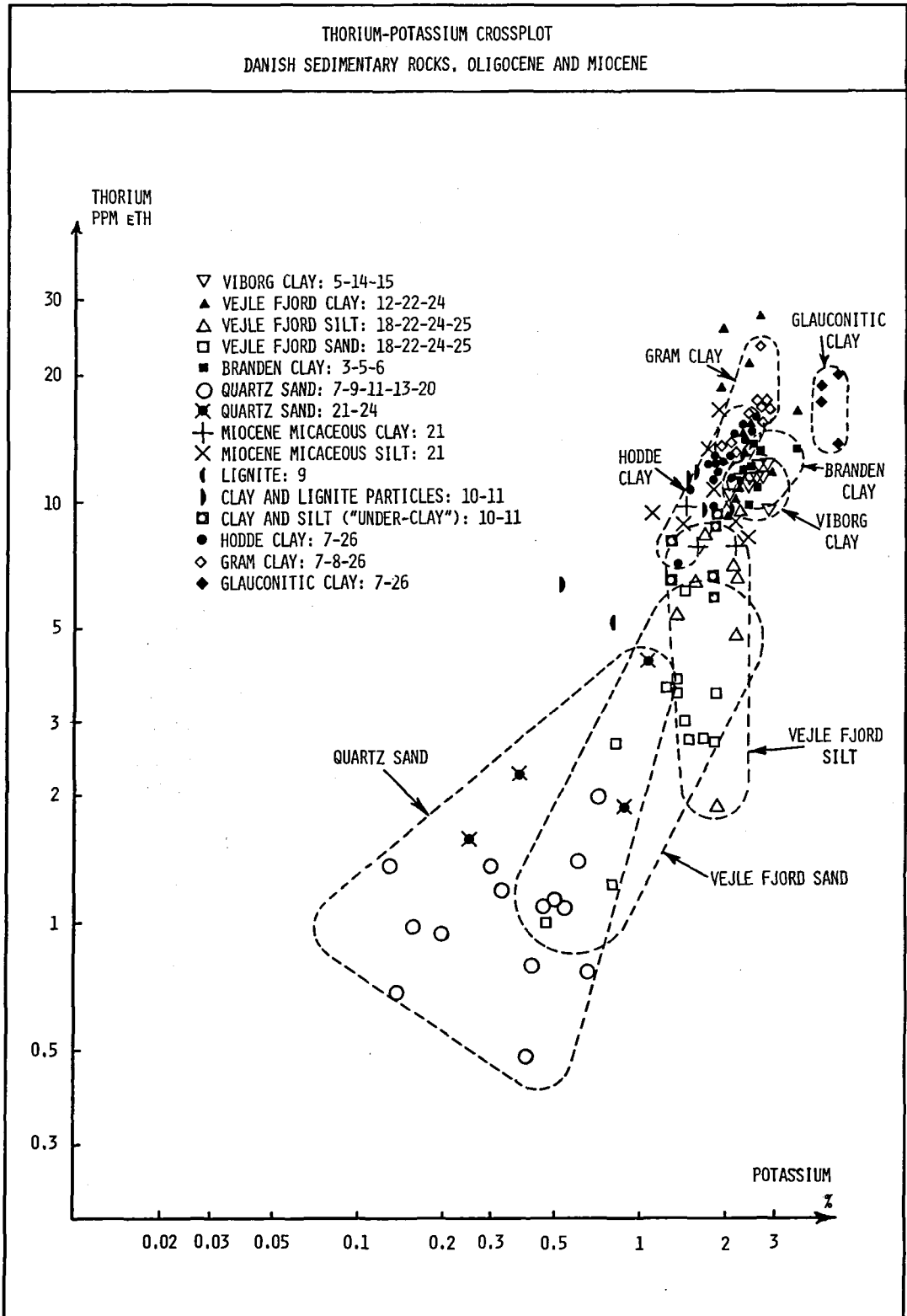


Fig. 7. Thorium-potassium cross-plot for Oligocene and Miocene sediments.

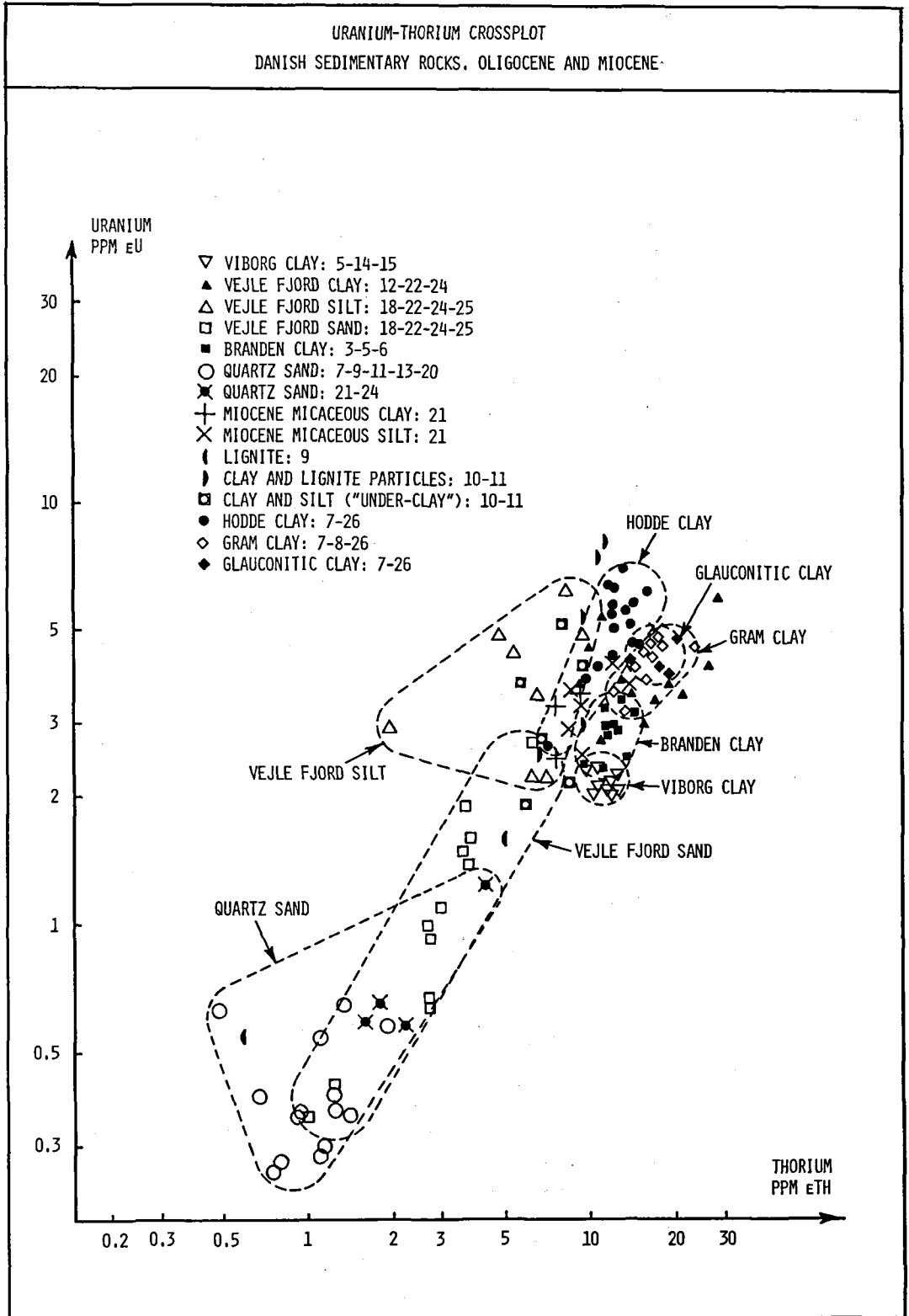


Fig. 8. Uranium-thorium cross-plot for Oligocene and Miocene sediments.

A closer examination of Mo-clay samples has shown that the Fur formation could be divided in an upper part and a lower part. The lower part contains higher concentrations of eU than the upper part, and the lower most parts of the Mo-clay exhibits the highest concentrations of eU we have found in any Danish formation.

The lower concentrations of eU found in the upper part of the Mo-clay could be due to the more frequent episodes of oxic conditions in the upper part of the Mo-clay (Pedersen, 1981) – or to a low “supply” of uranium during the deposition.

There are about 180 layers of volcanic ash in the Mo-clay. The ash layers have been given numbers from -39 to +140 (Bøggild 1918). Most of the layers from +1 to +140, which are contemporary to the upper part of the Mo-clay are of basaltic composition and contain moderate concentrations of the radioactive elements.

Ash layer number +19 has a dacitic composition and contain rather high concentrations of eTh and K (13 ppm; 2.5%) As this ash layer has a thickness of nearly 20 cm it might be used as a marker horizon in connection with SNG-logging.

Also ash layer number -17 of peralcaline composition might be a marker horizon for SNG-logging. The layer has a thickness of only about 4 cm, but it has very high concentrations of both eTh and eU.

## Younger Tertiary sediments

The figures 6 to 8 show the distribution of the radioactive elements in Danish sediments from Oligocene and Miocene. The concentrations vary much less than in the older sediments.

The lowest concentrations are found in Quartz Sand of Miocene age. The quartz grains have very low contents of radioactive elements but the sand also contains grains of heavy minerals and feldspars. Some of the heavy minerals contain thorium and uranium in rather high concentrations. (In samples of heavy minerals extracted from the Quartz Sand we have measured around 250 ppm thorium and 75 ppm uranium).

The feldspars include potassium feldspar, which is the site of the potassium measured in the samples of Quartz Sand.

The Vejle Fjord Sand is a micaceous sand,

composed mainly of quartz grains and overlap with the Quartz Sand in all three cross-plots.

Fine grains of heavy minerals are often deposited together with silt and thus contribute with thorium and uranium. This effect is seen in the Vejle Fjord Silt, which contains varying amounts of heavy minerals.

Most of the Danish clays of Miocene and Oligocene age overlap each other in the cross-plots. The Glauconitic Clay from the Gram Formation has, however, its “own” area in the U-K cross-plot and in the Th-K cross-plot due to its higher content of K.

The Vejle Fjord Clay has varying concentrations of both eTh, eU, and K and has not been assigned an area in the cross-plots.

According to the literature the concentrations of uranium may vary over a large interval in lignite, whereas the concentrations of K and eTh are negligible in pure lignite (Fertl 1979; Wedepohl 1978).

We have only had access to two samples of lignite (of Miocene age) both of which contain low concentrations of eU. One of the samples may be slightly “polluted” with clay as it contains small amounts of eTh and K.

The deposits of quartz sand in Jylland often contain strata of clay mixed with lignite particles. A few samples of this mixture have been measured and they contain eU, eTh, and K in varying amounts. The ratio between the concentrations of eU and eTh is rather high indicating a deposition in a reducing environment.

The same holds for the few samples of under-clay found together with the strata of clay and lignite particles.

## Example

Fig. 9 shows a SNG-log from a new water supply boring at Nebbegård (Sjælsø) north of Copenhagen. The log has been measured with the SNG-equipment built at our laboratory. The gamma-ray detector in the probe is a 3" × 3" NaI crystal.

The SNG-probe has been computer-calibrated and its accuracy has been checked with measurements in the laboratory and through logging in boreholes with known contents of radioactive elements. (Engell-Jensen 1982; DGU 1982;

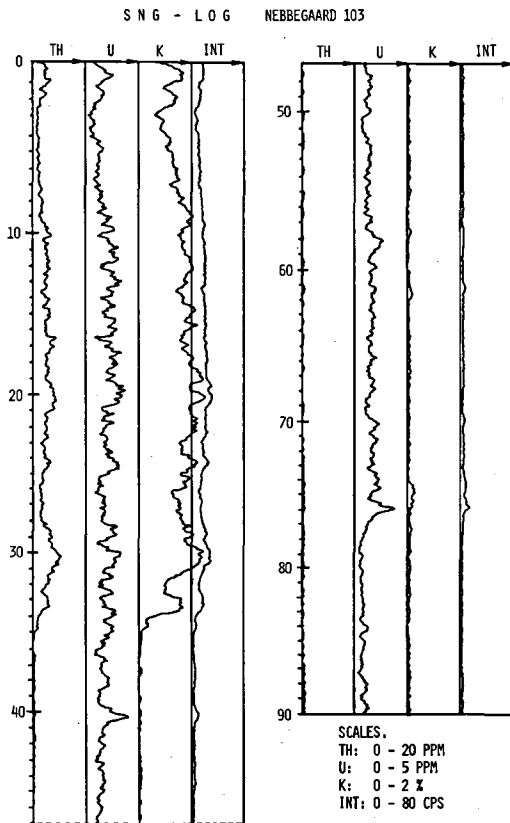


Fig. 9. SNG-log from a water-supply boring at Nebbegård north of Copenhagen. From 0 m to 34 m: Quarternary tills with varying contents of clay, silt and sand. Below 34 m: Danian calcarenite and flint with very low concentrations of thorium and potassium.

Jørgensen 1983; Korsbech, Jørgensen & Nielsen, in preparation).

The SNG-log shows the curves for the concentrations of eTh, eU, and K versus depth. Besides is shown the total count rate (integral) which is directly comparable to a usual natural gamma-ray log.

From the surface to 34 m is found Quarternary tills with varying contents of clay, silt, and sand. From 34 m to the bottom of the boring at 90 m is found Danian calcarenite and flint. Here the concentrations of eTh and K are very low.

At 40 m, 58 m, and 76 m are "peaks" of eU separating thick layers of rather homogeneous calcarenite and flint. The concentration of eU below 77 m is low (0.6–0.8 ppm) and higher between 41 m and 57 m (1.2–1.4 ppm) and between 59 m and 75 m (1.5–1.7 ppm).

A closer examination of the thorium curve tells that the concentration is slightly higher below 77 m than in the interval from 40 to 75 m. The difference may indicate a minor variation in the low content of clay.

SNG-logs from other borings some hundred meters away have similar appearance with uranium peaks separating thicker layers of calcarenite and flint with lower concentration of eU.

### Comparisons with other studies

Our measurements have shown that Danian limestone of bryozoan and coralline origin contain very low concentrations of both eTh and K, indicating a very low content of clay particles. The concentration of eU may be very low too (Fakse and Stevns) or higher (Hove). The very low concentrations of eU at Fakse and Stevns indicate a deposition in an oxidizing environment, whereas the limestone at Hove may have been deposited in a less oxidizing environment.

The observations concerning the carbonates are in agreement with the observations of other authors (Adams & Weaver, 1958 and Fertl, 1979).

The clay mineral content of some of our clayey samples has been measured and a reasonable correlation between the content of clay minerals and eTh was found, whereas the correlation with K was less obvious and no correlation existed for eU (Engstrøm, 1981).

These observations are in accordance with the experiences of others. Berstad & Dypvik (1982) e.g. found that thorium and potassium were associated with the clay fraction of Tertiary samples from the Central North Sea.

Heavy minerals often contain high concentrations of uranium and/or thorium (Adams & Weaver, 1958). As grains of heavy minerals are often concentrated in the silt fraction, the radioactivity of silt is influenced strongly by the composition of the heavy minerals. Monazite e.g. contains very high concentrations of thorium, whereas zircon in general is known to contain high concentrations of uranium.

We have found that both uranium and thorium are enriched in the silt fraction. Dypvik & Berstad (1982) found that uranium was enriched in the silt fraction. Different compositions of the

heavy minerals may explain the observed differences.

## Equilibrium of the radioactive decay chains

The gamma-rays which are measured in a borehole originate from potassium and from some of the radioactive decay products (daughters) of uranium and thorium. In general the radioactive chains following uranium and thorium are in equilibrium, i.e. neither uranium nor thorium, or any of their daughters, have been removed from – or added to – the sediments. Thus a measurement of the radioactive daughters is enough for a determination of the concentrations of uranium and thorium.

The radioactive thorium chain will reach an equilibrium within 30 years, which is a short period compared to a geological time scale, i.e. one has to expect equilibrium of the thorium chain in all geological samples.

The uranium chain needs about 1 mill. years in order to reach full equilibrium, and a disturbance of an equilibrium will not influence a SNG-log significantly until more than 20.000 years after the disturbance. In the laboratory one can, however, measure a possible non-equilibrium with no delay.

The question of an equilibrium or a non-equilibrium is of interest because uranium containing sediments which have been oxidized during or after the latest glaciation of Denmark may have lost some uranium – which could have been deposited in other sediments where reducing conditions prevailed.

In general we have found the uranium close to the equilibrium i.e. less than about 30% deviation from the equilibrium, but a few samples have lost uranium within the latest 100.000 years – and a few samples have gained uranium (50–400%). (Korsbech, in preparation).

The question of equilibrium or non-equilibrium within the uranium-chain does, however, not influence the interpretation of a SNG-log by means of the cross-plots. In the laboratory we – and most other investigators – have determined the uranium content of the samples by measuring the same gamma-rays as one measures in a borehole, i.e. the concentrations are given as

equivalent equilibrium uranium (eU). As a departure from equilibrium is the very same for the SNG-log as for the cross-plots the influence of a non-equilibrium is cancelled.

**Acknowledgements.** As we are engineers and not geologists, we have relied heavily on help and advice from people, who are more familiar with geology than we are. Ole Bjørsløv Nielsen and Erling Fuglsang, Department of Geology, University of Aarhus, have supplied several samples and have been of great help in the selection of formations and localities, which are included in this study. Birger Larsen, Birthe Dinesen and Kurt Klitten, Institute of applied Geology, Technical University of Denmark, have supplied other valuable samples.

Torben Pedersen, Gentofte Water-Supply Authority, has supplied us with more than 100 samples of Danian calcarenite and has given us access to several water-supply boreholes, where we have recorded SNG-logs. Henning Møller has done a major part of the laboratory work and Anne Borreby has typed the manuscript and improved the English text.

We wish to express our gratitude to all who have supported us in sampling, measurements, and preparation of this paper.

## Dansk sammendrag

Der er foretaget laboratorieundersøgelser af indholdet af de radioaktive grundstoffer thorium, uran og kalium i ca. 500 prøver af danske sedimentære bjergarter dækkende tiden fra Campan til Miocæn. Formålet med undersøgelsen har været at søge at opstille et grundlag for en lithostratigrafisk tolkning af SNG-logs (Spektral Naturlig Gamma-logs) optaget i borehuller med eller uden forerør.

Måleresultaterne præsenteres som diagrammer (cross-plots), hvor hver prøve er indtegnet i en position svarende til prøvens indhold af Th, U og K. Prøver af en bestemt type sediment bliver herved indtegnet som en samlet gruppe, der dækker et bestemt areal i diagrammet. Arealets størrelse er et mål for, hvor stor en variation der er mellem prøverne af den pågældende art.

Enkelte typer sedimenter indeholder så karakteristisk en sammensætning af radioaktive stoffer, at disse sedimenter entydigt kan identificeres på en SNG-log optaget med en kalibreret SNG-sonde ved hjælp af de fremstillede diagrammer (cross-plots). I andre tilfælde indeholder to eller flere typer sedimenter omtrent de samme koncentrationer af både Th, U og K, hvorfors de ikke entydigt kan identificeres af en SNG-log og diagrammerne. Andre typer information må da inddrages i tolkningen.

Variationer i indholdet af de radioaktive stoffer i en bestemt type aflejringer kan ved hjælp af en SNG-log i nogle tilfælde benyttes til korrelation mellem borehuller. I artiklen demonstreres et eksempel. SNG-logs optaget i nogle vandforsyningsboringer nord for København viser hvorledes indholdet af U – og til dels også Th – varierer med dybden i kalken på en karakteristisk måde, der kan følges fra borehul til borehul. En stratigrafisk korrelation i kalksandskalken, der ellers er vanskelig og tidskrævende, kan i et sådant område simpelt gennemføres ved optagelse af SNG-logs i borehullerne.

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