Pre-Quaternary rocks and sediments with a high level of radioactivity in Denmark

Peter Gravesen and Peter Roll Jakobsen

The pre-Quaternary sediments and rocks in Denmark generally have a low content of radioactive minerals and elements. Uranium, thorium and radium are built into mineral structures or are, for example, adsorbed on the surface of clay minerals, Fe-minerals or organic material.

Radon (^{222}Rn) is a radioactive noble insoluble gas with a half-life of 3.8 days. It belongs to the uranium (^{238}U) decay chain where radon is formed from radium (^{226}Ra). When Rn is formed by radioactive decay from Ra, the emanation process sends part of the radon produced into the pore spaces of rocks and soils. From here, the radon can enter and accumulate in buildings. The source of the radioactive materials in Danish sediments and rocks is primarily from weathered Precambrian crystalline rocks from Norway, Sweden, Finland and the Danish island of Bornholm. Physical and chemical weathering disintegrates these rocks and rivers transport the material into the Danish–Norwegian and Danish–Polish sedimentary basins.

Several studies have analysed and described the radioactive content of Danish sediments and crystalline rocks (e.g. Damkjær & Korsbech 1985, 1988; Gravesen *et al.* 1996, 1999; Gravesen & Jakobsen 2010) and investigations have demonstrated a relationship between sediments and rocks and Rn levels in Danish buildings (Andersen *et al*. 2001).

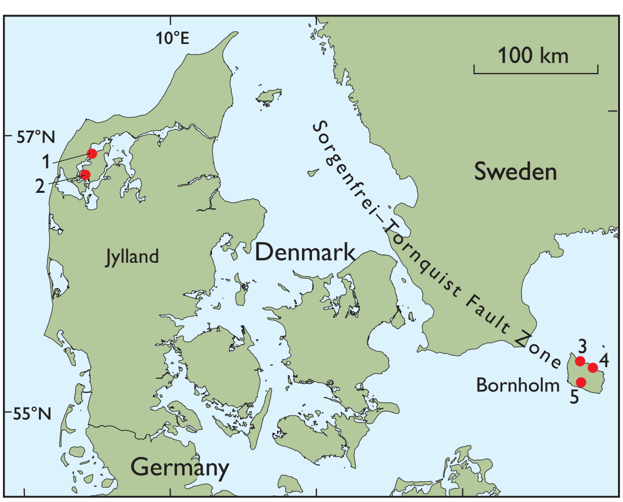

Fig. 1. Map of Denmark with the highest levels of radioactivity measured in surface rocks. **1**: Thisted. **2**: Erslev. **3**: Allinge. **4**: Tejn. **5**: Birkely.

This paper addresses the radioactive content of sediments and rocks with the highest radioactive levels in Denmark and the highest recorded radon emanations: Precambrian crystalline rocks on Bornholm and Late Paleocene clays in north-western Jylland (Fig 1). The data were collected by Gravesen *et al.* (1999) at the Geological Survey of Denmark and Greenland (GEUS) with the aim of characterising and mapping Rn in Danish rocks and sediments.

Methods and data

In Thisted, a 4 m long trench was dug, four shallow boreholes were drilled and older borehole data in GEUS' Jupiter database were studied. The trench was dug to a depth of *c.* 2 m into till deposits and limestone (Fig. 2) and the lithology, structures and macro-pores were described and samples collected. The four 5–6 m deep boreholes were drilled close to the trench. A total of 48 samples of the tills, clay and limestone were described and collected every 30 cm. The samples from the trench each weighed more than 500 g whereas the core samples weighed 300–500 g.

Studies and sampling were carried out at the Bornholm outcrops of granites and diabase at Allinge-Sandvig and Tejn (Fig. 1, loc. 3–4) at the northern end of the island and of the crystalline rocks at Birkely (near the farm of Vallensgård) to the south of Almindingen (Fig. 1, loc. 5). The lithology, structures, weathering, and faults and fractures

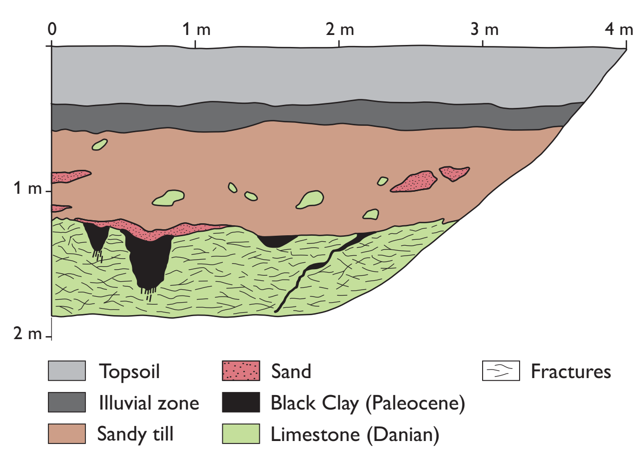

Fig. 2. Geological section in the trench from the eastern part of Thisted.

were described. A total of 10 samples of 500–1000 g were collected from the three localities.

The chemical and physical components of the matrix and the petrographical components of the clast material of the samples were analysed. The U concentration was measured by instrumental neutron activation analysis by Activation Laboratories Ltd, Canada. The Ra content was measured with a germanium detector at the National Institute of Radiation Protection, Denmark. The Rn emanation rate was determined using the closed-chamber method using ZnS(Ag) scintillation cells at the Risø National Laboratory, Technical University of Denmark.

Relationship between Danian and Late Paleocene deposits in Thisted

The distribution of Danian – Late Paleocene (65.5–55.8 Ma) deposits in north-western Jylland in the area of Mors and Thisted is related to two salt diapirs, the Erslev salt diapir (Pedersen *et al.* 2013) and the Thisted salt dome (Hansen & Håkansson 1980). Sediment distribution around the two salt structures is similar at the pre-Quaternary surface with Maastrichtian chalk in the middle surrounded by Danian limestone and with Palaeogene clays bordering the limestone. The layers are generally inclined away from the centre of the salt structures.

Different aspects of the radioactive components in the areas were investigated by Damkjær & Korsbech (1985) and Gravesen *et al.* (1996). The Quaternary cover above the Danian limestone is rather thin (0–2 m) in the Thisted area. A topsoil layer overlies Weichselian glacial sandy tills, clayey tills, and just above the limestone is a strongly calcareous till (about 60–80% $CaCO_3$). These tills contain vertical fractures at 5–10 cm intervals.

The Late Danian limestone is partly cemented and can be classified as a calcilutitic or calcarenitic limestone according to grain size. The trenches demonstrated more complex geological structures than expected (Fig. 2). The top surface of the Danian limestone was eroded and brecciated during the Late Paleocene and cut by horizontal and vertical fractures with pronounced karst features and inclined fractures filled with black structureless Paleocene clay which contains Selandian dinoflagellate cysts. The clay also contains limestone clasts and is probably redeposited. At the bottom of the karst structures, several clay-filled fractures reach at least 10–15 cm downwards (Fig. 2). In a few shallow boreholes at Thisted, more than 50 cm of black clay has been encountered indicating non-eroded remnants of the black clay, and black clay material is also enclosed in the till at some locations.

The content of radioactive material in the Thisted sediments is presented in Table 1 (Data from Damkjær & Korsbech 1988 and Gravesen *et al.* 1999). The U, Ra and Rn values for the Danian sediments are comparable with levels for these sediments in other parts of the country (Damkjær & Korsbech 1985) but the high value for the radon emanation of the black clay can only be compared with levels of Paleocene black clay from the Erslev area which has even higher values (Table 1). High levels of radon emanations are also known from Cambrian–Ordovician alum shale: 16 atoms/kg/s, Eocene diatomites: 22 atoms/kg/s, Miocene U-bearing heavy sand: 52 atoms/kg/s, and Miocene black clay: 38.9 atoms/kg/s (Damkjær & Korsbech 1985).

Discussion

Karst in Danish limestone and chalk is found in many areas of northern Jylland but the occurrence in the Thisted trench is noteworthy due to its radioactivity. Karst is formed by acidic water percolating through fractures in the limestone and dissolving parts of it. The erosion of the pre-Quaternary surface started at the inversion and uplift of the Sorgenfrei–Tornquist zone at the end of Cretaceous–Paleocene time followed by Cenozoic sub-areal erosion (Stenestad 2006). The erosion had removed the Late Paleocene clay from most of the Thisted area and redeposited part of the material in the shallow karst holes in the limestone.

In a search for the origin of Rn in buildings Damkjær & Korsbech (1988) investigated sediments from the Erslev area and suggested that redeposited black clay in tills was the source of the high levels of radon. However, the results showed that the high values were in the limestone areas. The later investigation of Andersen *et al.* (2001) demon-

Table 1. Measured radioactive components

Locality	Lithology and age	Uranium ppm	Radium Bq/kg	Radon Atoms/kg/s
Thisted	Weichselian sandy till	0.8–1.9	18.7–24.7	7.5–10.4
	Late Paleocene clay	–	–	42.4
	Danian limestone	< 0.6	2.8–3.3	0.12–0.8
Erslev	Paleocene clay	2.72–26.3	38–300	11.2–130
Allinge	Hammer granite (weathered)	4.2	86.9	–
	Hammer granite	3.0	63.5	–
Tejn	Vang granite	3.8	66.0	–
Birkely	Almindingen granite (weathered)	3.4	41–51	9.6
	Almindingen granite	4.3	50.7	8.8
	Diabase (weathered)	4.2	104.2	39.6
	Clay: weathered diabase	8.5	–	–

strated relatively high levels of Rn in buildings sitting on limestone in Thisted although the radioactivity of limestones at both localities was among the lowest in Danish sediments. The present investigation from Thisted based on the trench and shallow boreholes shows that the high levels of Rn emanation from isolated or redeposited Late Paleocene black clays can probably be the source of the high Rn levels in the buildings in this region. In Sussex, England, high Rn levels in houses are also partly caused by redeposited material on top of low-radioactive chalk (Killip 2004).

Precambrian basement rocks: radioactivity and weathering

Bornholm is situated in the Sorgenfrei–Tornquist Fault Zone south of Sweden (Fig. 1). The Precambrian basement of northern and eastern Bornholm consists of granitic and gneissic rocks which contain abundant leucogranitic bodies, pegmatites and aplites, besides more than 250 mafic dykes that cut these crystalline rocks.

The Kampeløkke Å locality at Allinge consists of medium-grained Hammer granite with a *c.* 20 cm thick crust of weathered granite (Fig. 3). The Hammer granite comprises 41% K-feldspar, 18% plagioclase, 33% quartz, 1% hornblende, 4% biotite, and accessory Ti-magnetite, apatite, epidote, allanite and fluorite (Micheelsen 1961). Large crystals of black gadolinite, a REE-Fe-Be silicate mineral, are found in the area with abundant small red spots of Fe_2O_3 on its crystal surfaces. The granite is cut by vertical and horizontal fractures. The content of radioactive compounds is seen in Table 1. In the Hammer granite the U content is between 3.0 and 4.2 ppm and the Ra content is between 63.5 and 86.9 Bq/kg.

The Møllebæk locality at Tejn consists of coarse-grained Vang granite with very coarse-grained pegmatites covered by 10–20 cm thick weathered granite. The granite contains vertical and horizontal fractures. The Vang granite is composed of 33% K-feldspar, 22% plagioclase, 27% quartz, 5% hornblende, 6% biotite, 3% Ti-magnetite, 1% titanite, 1% apatite and subordinate allanite (Micheelsen 1961). The pegmatites have only a low content of dark minerals (averaging 1%) but 45–60% K-feldspar and 30–40% quartz. The content of radioactive components is comparable to that of the Hammer granite.

The Birkely locality (1½ km north of the farm Vallensgård) is a small quarry with medium-grained Almindingen granite. The mineralogical composition is nearly the same as for the Hammer granite. Figure 4 shows a section with a strongly fractured and faulted part with several partly weathered olivine diabase dykes and weathered granite.

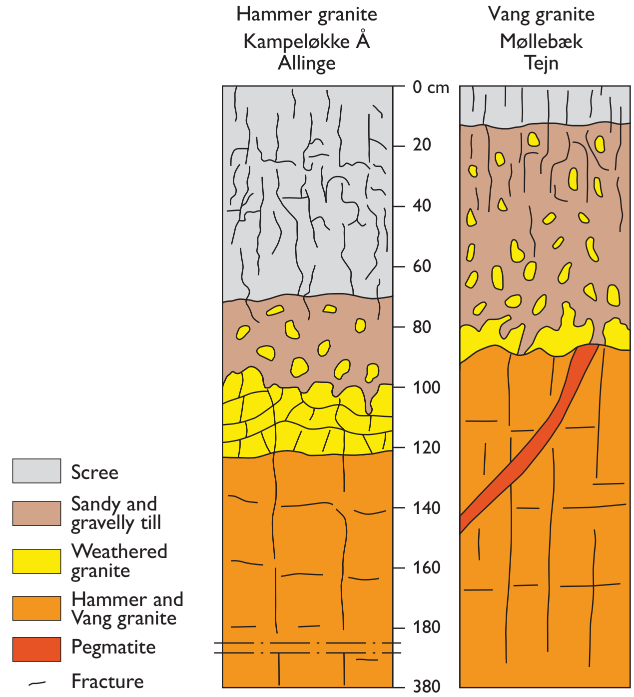

Fig. 3. Geological sections from Allinge and Tejn.

These faults and fractures are orientated WNW–ESE, the same direction as the major fault between the crystalline rocks and the younger sediments (Micheelsen 1961), and the faults have slickensides demonstrating movement.

The diabase dykes mainly contain olivine and pyroxene but biotite, epidote and hornblende also occur (Callisen 1934). Alteration of these minerals produced serpentine, chlorite and calcite. The diabase dykes are orientated NW–SE as are many other diabase dykes and faults in these rocks. Along its contacts with weathered granite, the diabase is altered to green clayey material of chlorite or serpertine (Fig. 4). The rocks in the area are partly weathered and Fe-bearing minerals are oxidised to yellow-brown, clayey iron-rich weathering products on the fracture surfaces. The content of radioactive components is shown in Table 1.

Discussion

In this study, the U content of the Hammer, Almindingen and Vang granites was found to be below 5 ppm, but higher values (6–16 ppm) have previously been recorded (Johansson *et al.* 2016). The olivine diabase dyke yielded a comparable U content, about 4.2 ppm. The granites and diabases have high levels of U, Ra and emanations of Rn, and examples of weathered granites and diabases with higher levels are found. Some of these rocks are among those with the highest radon emanations known in Denmark. The

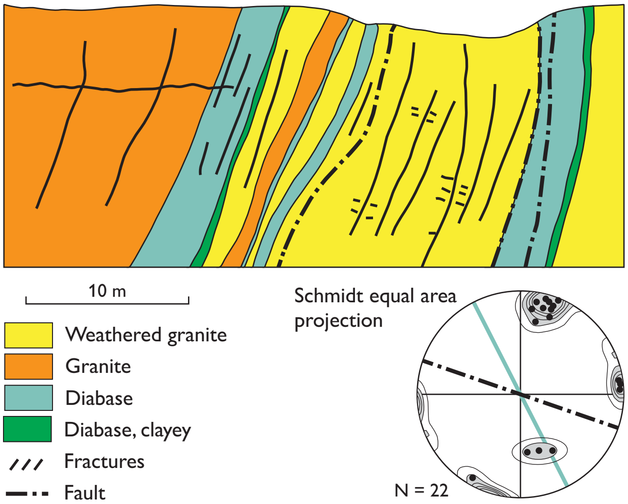

Fig. 4. Geological section from the Birkely quarry, south of Almindingen, Bornholm. The orientation of faults and fractures, and diabase sheets (green) are shown in the diagram.

weathered diabase yields Rn emanation levels comparable to the Paleocene clay from Thisted.

According to leaching investigations of Pliler & Adams (1962), granitic rocks become depleted in U during the first phase of acid chemical weathering, but during the subsequent weathering phases, the top of the weathered rocks become altered and now predominantly consist of resistant U-bearing minerals. The weathering of the ground surface and shallow fractures of the rocks is mainly due to hydrolysis by percolating surface water and fluctuating low temperatures.

The mafic minerals in diabases, such as hornblende, pyroxene and biotite, can be altered to green chlorite or serpentine and eventually form a clayey material hosting the U.

The crystalline rocks are the primary source of radon in buildings on northern Bornholm, and especially where cellar walls consist of unweathered or weathered rocks the Rn concentrations in the buildings can be high (Andersen *et al.* 2001).

Conclusions

The black Paleocene clay on Danian limestone in Thisted and the crystalline rocks on Bornholm are among the Danish deposits with the highest contents of U and Ra, and they also have the highest Rn emanation rates. Other sediments at this emanation level are fine-grained Cambrian–Ordovician alum shale, Eocene diatomites and Miocene black and brown clays and sand. Most of these sediments, like the Thisted clay deposits, have limited distribution and are found below thick Quaternary layers and can be difficult to locate.

Acknowledgements

The Danish Ministry of Health is thanked for financial support.

References

Andersen, C.E., Ulbak, K., Damkjær, A. & Gravesen, P. 2001: Radon i danske boliger. Kortlægning af lands-, amts- og kommuneværdier, 74 pp. Copenhagen: Sundhedsstyrelsen.

Callisen, K. 1934: Das Grundgebirge von Bornholm. Geological Survey of Denmark, II. Række 50, 266 pp.

Damkjær, A. & Korsbech, U. 1985: Measurement of the emanation of radon-222 from Danish soils. The Science of the Total Environment **45**, 343–350.

Damkjær, A. & Korsbech, U. 1988: A search for correlation between local geology and indoor radon concentration. Radiation Protection Dosimetry **24**, 51–54.

Gravesen, P. & Jakobsen, P.R. 2010: Radon content in Danish till deposits: relationship with redox conditions and age. Geological Survey of Denmark and Greenland Bulletin **20**, 39–42.

Gravesen, P., Jakobsen, P.R. & Kelstrup, N. 1996: Radon i danske jordarter II, undersøgelser og konklusioner. Danmarks og Grønlands Geologiske Undersøgelse Rapport **1996/78**, 113 pp.

Gravesen, P., Jakobsen, P.R., Kelstrup, N. & Ernstsen, V. 1999: Kortlægning af radon i danske jordarter 1. Indsamling af grunddata. Danmarks og Grønlands Geologiske Undersøgelse Rapport **1999/81**, 58 pp.

Hansen, J.M. & Håkansson, E. 1980: Thistedstrukturens geologi – et "neotektonisk" skoleeksempel. Dansk geologisk Forening, Årsskrift for **1979**, 1–9.

Johansson, Å., Waight, T. Andersen, T. & Simonsen, S.L. 2016: Geochemistry and petrogenesis of Mesoproterozoic A-type granitoids from the Danish island of Bornholm, southern Fennoscandia. Lithos **244**, 94–108, *http://dx.doi.org/ 10.1016/j.lithos.2015.11.031*

Killip, I.R. 2004: Radon hazard and risk in Sussex, England and the factors affecting radon levels in dwellings in chalk terrain. Radiation Protection Dosimetry **113**, 99–107.

Micheelsen, H.I. 1961: Bornholms grundfjæld. Bulletin Geological Society of Denmark **14**, 308–349.

Pedersen, S.A.S., Jakobsen, P.R., Tougaard, L. & Gravesen, P. 2013: Geological map of Denmark 1:50 000 – map sheet Mors, NW Denmark. Geological Survey of Denmark and Greenland Bulletin **28**, 29–32.

Pliler, R. & Adams, J.A.S. 1962: The distribution of thorium and uranium in a Pennsylvanian weathering profile. Geochimica et Cosmochimica Acta **26**, 1137–1146.

Stenestad, E. 2006: Fluviokarst in the top of the Maastrichtian chalk at Rørdal, northern Jutland, Denmark. Bulletin of the Geological Society of Denmark **53**, 93–110.

Authors' address

Geological Survey of Denmark and Greenland, Øster Voldgade 10, DK-1350 Copenhagen K, Denmark. E-mail: *pg@geus.dk*