Palynology of Early Cretaceous (Barremian to Aptian) hydrocarbon (methane) seep carbonates and associated mudstones, Wollaston Forland, Northeast Greenland

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ABSTRACT. Palynostratigraphic and palynofacies analysis have been performed on hydrocarbon seep carbonate, carbonate nodule and mudstone samples from the Early Cretaceous Kuhnpasset Beds in the Kuhnpasset area of Wollaston Forland, Northeast Greenland. Three informal palynostratigraphic zones have been defined based on dinoflagellate cyst occurrences. The zones range from ?early Barremian to early Aptian in age, and correlate with previously defined dinoflagellate cyst zones in Northeast Greenland. These zones indicate hydrocarbon seepage in Kuhnpasset spanned the ?early–late Barremian age range, but did not continue into the Aptian, and thus seep activity continued for as much as three million years. Palynofacies analysis is used to interpret the depositional environment of the Kuhnpasset Beds and indicate that the hydrocarbon seeps and associated mudstones from this sedimentary sequence were deposited in a proximal shelf setting.

KEYWORDS: Palynostratigraphy, dinoflagellate cysts, palynofacies, hydrocarbon seep carbonates, Early Cretaceous, Northeast Greenland

INTRODUCTION

Hydrocarbon seeps are places on the continental margins where fluids rich in hydrocarbons, principally methane, flow on to the seafloor (Sibuet and Olu, 1998). First discovered in 1984, they are now found in all the World’s oceans. Hydrocarbon seeps support highly unusual biotic communities, which, together with the similarly structured hydrothermal vent communities, have altered our view of life in the deep sea, in part because the primary energy source for these ecosystems is not solar, but geochemical (Levin, 2005). In the shallow subsurface at seep sites methane is utilized by a consortium of methanotrophic archaea and sulphate-reducing bacteria in the anaerobic oxidation of methane reaction (e.g. Hinrichs et al., 1999; Boetius et al., 2000; Reitner et al., 2005), leading to the supersaturation of pore fluids with respect to carbonate ions and resulting in the formation of distinctive authigenic carbonate deposits with multi-phase carbonate cements and very negative $\delta^{13}$C values (e.g. Ritter et al., 1987; Aloisi et al., 2000; Naehr et al., 2007; Haas et al., 2010). Seep carbonates...
have a wide variety of morphologies (e.g. nodules, tubular/tabular concretions, cemented breccias and pavements) and sizes (e.g. Campbell, 2006), and because of rapid precipitation commonly contain well-preserved macrofossils and microfossils, including palynomorphs (e.g. Kiel et al., 2013; Dalseg et al., 2016a, b).

The Mesozoic is a crucial time in the evolutionary history of seep faunas, with a change from brachiopod dominated communities of the Triassic, Jurassic and Early Cretaceous to bivalve dominated communities of the Late Cretaceous (Sandy, 2010). Palaeobathymetric controls on Mesozoic seep faunas occurred in seep communities in the Cretaceous, as in modern seeps, with the number of obligate species decreasing from the slope and deep shelf to the shallow shelves (Kiel, 2010). Compared to the Cenozoic, the Mesozoic fossil record of hydrocarbon seep communities is relatively sparse and comes largely from the circum-Pacific area (e.g. Japan and Western USA) and sites that were in the Tethys Ocean (principally Southern to Eastern Europe; Kiel et al., 2008; Kaim et al., 2013). Four areas of Jurassic to Cretaceous aged hydrocarbon seepage are known from the present-day Arctic region, including two sites in Northeast Greenland: one Barremian (Kuhnpasset, Wollaston Forland; Kelly et al., 2000) and one Campanian (Leitch Bjerg, Geographical Society Ø); Svalbard (Jurassic–Cretaceous boundary; Hammer et al., 2011); Arctic Canada (Albian of Prince Patrick Island and Ellef Ringnes Island; Beauchamp and Savard, 1992; Williscroft et al., 2017) and Novaya Zemlya (three ages of seepage: late Oxfordian–early Kimmeridgian, late Tithonian and latest Berriasian–early Valanginian; Hryniewicz et al., 2015). In the Mesozoic, the latter three areas were part of the Boreal Ocean, which was a relatively isolated sea with limited marine connections with the Tethys and ancient Pacific (Panthalassic) Oceans (Zakharov et al., 2002). The Northeast Greenland seeps occurred in a more southerly position in a narrow seaway that linked the Boreal Ocean to the Tethys, through Northwest Europe.

The seeps in Kuhnpasset have been described by Kelly et al. (2000) and comprise over 30 exposed carbonate mounds (27 individually numbered) on the eastern side of Kuhnpasset, below the western flank of Aucellabjerget Mountain (Fig. 1). All but one of these are concentrated along a ridge in an area of ~1 km² (Kelly et al., 2000: fig. 4). The individual seep carbonates are subcircular to suboval in plan view, with relatively flat bases, where these can be seen. They vary in size from 1 to 3 m in diameter and are up to 1.8 m in height. The host rocks to the carbonate bodies are silty to sandy mudstones, and subordinate thin sandstone beds (Fig. 2). Kelly et al. (2000) erected the name Kuhnpasset Beds for this sequence and suggested a late Barremian age for it, based on the occurrence of the ammonite genera Audouliceras, Epicheloniceras and Sanmartinoceras, and the presence of dinoflagellate cysts indicative of the Pseudoecerium toveae Subzone (Nøhr-Hansen, 1993). Subsequently, Bjerager et al. (2020) incorporated the Kuhnpasset Beds into their Stratumbjerg Formation of the Brorson Halvø Group. Bjerager et al. (2020) suggested that the Stratumbjerg Formation was predominantly deposited in slope and basin-floor settings, with local passive infill of submarine gullies and shallow marine deposition. The latter facies is represented in the Kuhnpasset-Aucellabjerget area by the cross-bedded sandstones that overly the seep bearing mudstone sequence (Fig. 2). Bjerager et al. (2020) named these sandstones the Aucellabjerget Member, and this is thought...
to be late early Aptian in age, based on dinoflagellates (Nøhr-Hansen et al., 2020) and the occurrence of the ammonite genus *Deshayesites* (Kelly et al., 2000).

In this paper we present a detailed investigation of the palynofloras from the seep-bearing sedimentary sequence from Kuhnpasset, including, for the first time, from the seep carbonates themselves. Our aims are to better refine the age of the seep carbonates and the duration of hydrocarbon seepage in the area by comparing local palynostratigraphic zones with established Cretaceous dinoflagellate cyst zonations from Greenland and other areas from the Boreal realm. In addition, we perform a palynofacies analysis to interpret the depositional environment during the formation of the seeps in Kuhnpasset.

Figure 2. Lithological log with sample levels indicated (red boxes). Lithostratigraphy from Bjerager et al. (2020)
MATERIALS AND METHODS

The material for this study was collected during fieldwork to Kuhnpasset between the 2nd and 12th August 2019 by Crispin T.S. Little, Hans Arne Nakrem, and Simon R.A. Kelly under Prospecting License No. 2017/15 from the Ministry of Industry, Energy and Research, Government of Greenland. A sedimentary log was made from seep carbonate number 27 of Kelly et al. (2000) at the base, to the sandstones of the Aucellabjerg Member at the top of the section (Fig. 2). We selected 13 mudstone samples from this section, one carbonate nodule and six seep carbonates for subsequent analysis (Supplementary File 1). Between 700 and 2000 g of carbonate nodule and seep carbonate samples were treated with 10–15% acetic acid at the University of Oslo. After five days, the acid had neutralized and the resulting residues were sieved. The <63 μm fraction was retained and sent with the mudstone samples (approximately 100 g per sample) to Applied Petroleum Technology A/S (APT), Oslo for standard processing onto palynological slides. These slides were studied by Emil Bang at the University of Oslo using a Leica DMLP microscope. Photomicrographs of palynomorphs were taken with a Leica camera MC170HD and Leica Application Suite v. 3.4.0 – LAS software. The images were stacked in Helicon Focus v.7.0 and edited in ACDSee Ultimate 8 and Adobe Photoshop CS6 software. The dinoflagellate cysts in the slides were identified based on shape, location of archaeopyle, and shape of processes (if present) using works by Davey (1982), Heilmann-Clausen and Birkelund (1987), Nøhr-Hansen (1993), Nøhr-Hansen et al. (2020) and Śliwińska et al. (2020).

Dinoflagellate cyst abundance was calculated as individuals of each taxon per total individuals in each sample. The abundances were sorted into four abundance groups: Rare (0–0.99%), Common (1.0–9.9%), Abundant (10.0–24.9%) and Dominant (25.0–100%). For each sample 300 specimens were counted. For dinoflagellate cyst diversity indices Simpson 1-D index, Shannon index (Shannon-Wiener index) and Fisher-α index were calculated. The calculations were performed using PAST software ver. 4.06 (Hammer et al., 2001; Hammer, 2021). For the palynofacies analysis, approximately 300 organic particles were counted for each sample. Ideally 500 organic particles should be counted (Tyson, 1995), but many of our slides contained fewer than this, therefore we consider 300 organic particles to be adequate for our analyses. The counts were divided into the following groups: structured organic matter (spores, pollen grains, dinoflagellate cysts, cuticles, coaly phytoclasts, woody phytoclasts, other plant debris) and structureless organic matter (amorphous organic matter; AOM). Cuticles, coaly phytoclasts, woody phytoclasts and other plant debris are furthermore grouped in phytoclasts.

Figured specimens (Pl. 1) are curated in the Natural History Museum, University of Oslo, abbreviated PMO (former Paleontologisk Museum Oslo). The taxonomy used here follows that of Williams et al. (2017).

RESULTS

The preservation of palynomorphs in the samples was very good throughout all samples, with seemingly no difference between the seep carbonate and mudstone samples. However, palynomorphs in the uppermost two samples were less well preserved than those in the other samples. This material also had a darker color (TAI 2/2+) than the other samples, which generally had a bright yellow coloration (TAI 1/1+). In total, 99 different dinoflagellate cyst taxa were identified from the carbonate and mudstone samples (Pl. 1, Supplementary File 2). This included 41 genera, 54 species, one affinity (aff.) to a species, four confers (cf.) to a species and five confers to a genus; four were primarily identified to genus level (Gonyaulacysta sp., Kiokansium sp., Spiniferites sp., Subtilisphaera sp.). From the mudstone samples a total of 84 taxa were identified, including 48 which were identified to species level. The remaining taxa were placed in open nomenclature. For the carbonate samples a total of 69 taxa were identified, 39 to species level, 30 in open nomenclature. The terrestrial palynomorphs observed in the samples were not identified more specifically than general groups of spores or pollen. In addition, some freshwater algae were observed. Only two observed palynomorphs were identified as being reworked. These were a Jurassic dinoflagellate cysts of the genus Rigaudella Below 1982 (sensu Nøhr-Hansen, 1993) in Seep 3A (90.8 m level), and the Permian bisaccate pollen Protohaploxypinus sp. in the carbonate nodule sample (78.3 m level).

PALYNOSTRATIGRAPHIC ANALYSIS

The stratigraphic distribution of identified dinoflagellate cyst taxa is presented in Figs 3 and 4. Using these stratigraphic ranges we divide our Kuhnpasset section into three informal zones based on first occurrences (FO) and last occurrences (LO) of selected dinoflagellate cyst species (Fig. 4). An informal zoning is preferred as the materials in this study were not collected with regular sampling intervals; rather, sampling was adjusted to the seep

1 Supplementary File 1: Sample list and weights
2 Supplementary File 2: Dinoflagellate cyst taxa counts per sample.
carbonate and adjacent mudstone intervals. Our informal zones are defined as follows, from base upwards:

**Zone 1.** This encompasses all samples from Seep 27 up to Seep 26, at 97.6 m in the section. The upper zone boundary is defined at the LO of *Hystrichosphaerina schindewolfii* and the FO of *Palaeoperidinium cretaceum*. The lower boundary of the zone is left open. *Batioladinium exiguum*, *Endoscrinium campanula* and *Hystrichosphaeridium arborispinum* have their first and last occurrences within this zone.

**Zone 2.** This spans the interval from Seep 26 to the uppermost mudstone sample above New Seep 2019, at 104.6 m in the section. The zone is transitional with stepwise LOs of different species through the zone. The lower boundary is placed at the LO of *Hystrichosphaerina schindewolfii*, and the upper boundary at the LO of *Pseudoceratium anaphrissum*, *Chlamydophorella nyei* and *Gardodinium trabeculosum*. The zone boundaries coincide with the FO and LO of *Palaeoperidinium cretaceum*. The oldest possible age for zone 1 is early Barremian as *Pseudoceratium anaphrissum* is present throughout the entire zone. According to Nøhr-Hansen (1993), *P. anaphrissum* has its uppermost occurrence in the early Barremian *Pseudoceratium anaphrissum* Subzone. In our study *Pseudoceratium toveae* occurs as a dominant species at 86.6 m. This may correlate with the acme of this species recorded in the *Pseudoceratium toveae* Subzone by Nøhr-Hansen (1993). However, this subzone was defined above the LO of *P. anaphrissum* and dated as late Barremian (Nøhr-Hansen, 1993). If so, it suggests that our

**Zone 3.** This comprises the two uppermost samples in the section (115.0 and 116.3 m levels). The lower boundary of the zone is defined by the LO of *Chlamydophorella nyei*, *Gardodinium trabeculosum* and *Palaeoperidinium cretaceum*. No upper boundary is defined here. Our informal zones 1–3 can be roughly correlated with the Northeast Greenland dinoflagellate zones defined by Nøhr-Hansen (1993) and Nøhr-Hansen et al. (2020).
Figure 4. Dinoflagellate cyst species range chart, with abundance data and informal zones 1–3.
zone 1 is of early-late Barremian age. It is also possible that *P. anaphrissum* is reworked, but the common occurrence of this species at the top of subzone 1 makes this unlikely. Our zones 1 and 2 could represent the transition from the *P. anaphrissum* Subzone to the *P. toveae* Subzone, but a more detailed investigation is needed to address this question. Within our zone 2, *Batioladinium longicornutum* has its LO, suggesting that this zone has a latest Barremian age. This is based on correlation with the *Batioladinium longicornutum* zone redefined by Nøhr-Hansen et al. (2020), which is recognized on the basis of the FO and LO of *Batioladinium longicornutum*. With *Odontochitina nuda* present in our zone 3, and the LO of *Batioladinium longicornutum* in our zone 2, zone 3 is likely of early Aptian age. This is based on correlation with the *Odontochitina nuda* Zone of early Aptian age in the work of Nøhr-Hansen (1993), the lower boundary of which is defined on the basis of the LO of *Batioladinium longicornutum* and the upper boundary at the LO of *Odontochitina nuda*. Thus, seepage in Kuhnpasset spanned the early–late Barremian age range, but did not continue into the Aptian. This extends the duration for seepage suggested by Kelly et al. (2000) from the late to the early Barremian, and indicates that there was seep activity over some extended period of time, possibly as much as three million years, beginning with the formation of Seep 27 and ending at New Seep 2019. Saying that, the majority of seepage would have occurred over a shorter period of time, perhaps confined to the late Barremian, as the majority of seep deposits are confined to 20 m of the Kuhnpasset section (Fig. 2).

In addition to the key species listed above, the occurrence of *Pseudoceratium anaphrissum* (Late Hauterivian–early late Barremian) and *Vesperopsis longicornis* (late Barremian–early Albian) in our material is also of biostratigraphic significance. In our study we found *Batioladinium longicornutum* had an LO before that of *Pseudoceratium anaphrissum*. This contrasts with the Northeast Greenland dinoflagellate cyst biostratigraphy of Nøhr-Hansen (1993) and Nøhr-Hansen et al. (2020), where the late Barremian *Pseudoceratium toveae* Subzone is defined from the LO of *Pseudoceratium anaphrissum* to the LO of *Batioladinium longicornutum*. An explanation for this difference could be that the LO of *Batioladinium longicornutum* is within the upper part of the section, from which we did not collect samples for palynomorph study (between 104.8 and 115.0 m), or because of reworking, as we found reworked Permian and Jurassic palynomorphs in some of our samples.

Although not the subject of this study, the Kuhnpasset Beds have a rich ammonite fauna, dominated by lytoceratids, but also including aconiceratids, cheloniceratids and heteromorphs. These are currently undescribed, but based on preliminary investigation, compare with some of Rosenkrantz’s (1934) material from Kuhn Ø and Frebold’s (1935) from Storre Koldewey, both of stated Aptian age. However, the earliest reliably dated ammonite which we discovered in 2019 at Kuhnpasset was a specimen of *Deshayesites boegvadi* Rosenkrantz (1934), from about 2 m above the base of the Auclellabjerg Member (122 m on Fig. 2). This ammonite was taken as an indicator for the mid early Aptian in Hold with Hope (Kelly and Whitham, 1999) and confirms an early Aptian age for the strata above the Kuhnpasset seeps. Further work to reconcile ammonite and dinoflagellate cyst biostratigraphy in Kuhnpasset specifically and Northeast Greenland in general would be beneficial.

**Dinoflagellate Cyst Diversity**

Within our zone 1, the number of taxa per sample varies between 27 and 46, in zone 2, between 29 and 37. In zone 3, the number varies between 19 and 20. Our diversity data corresponds well with the observations of Nøhr-Hansen (1993) who reported similar diversities in the Barremian samples, and a lower diversity in the late early Aptian – early Albian samples. Note that Nøhr-Hansen (1993) recorded diversity as the number of species, whereas here diversity is recorded as the total number of taxa, therefore a direct comparison of the actual number of species is not possible.

Our statistical analyses indicate that in the mudstone samples the Simpson 1-D index averages at 0.9 and varies between 0.9 and 1.0. In the carbonates the Simpson 1-D also averages at 0.9, with no variation. The Shannon H index in the carbonate samples has the lowest value at 2.9 and highest at 3.2, averaging at 3.0. This is slightly lower than the mudstone samples averaging at 3.1. The
mudstone samples also have larger outliers, with lowest $H$ at 2.4 and highest at 3.5. The Fisher-$\alpha$ averages at 10.8 in the mudstone samples and 9.8 in the carbonates. The lowest value for Fisher-$\alpha$ in the mudstones is 5.8 and 8.9 for the carbonates. The highest value for the latter is 11.1 while for the mudstones it is 15.0. The variation between sample types and diversity indices is presented in Fig. 5. In general, our recorded diversity indices show a relatively stable Simpson 1-D value for all samples, whereas the Fisher-$\alpha$ and Shannon $H$ values have larger variation. According to this, it seems that the dinoflagellate cyst diversity in general is quite high, but with a slightly lower diversity in the seep carbonates. This could be the result of a changing environment when hydrocarbon seepage took place, which impacted the water column around the area of seepage, or due to differential preservation in the sediment due to the anaerobic oxidation of methane producing chemical gradients not conducive to the preservation of organic matter. There is also a decreasing trend in diversity in the two uppermost mudstone samples. This may be partly due to the poorer preservation observed in these samples.

### PALYNOFACIES ANALYSIS

Results of the palynofacies analyses are presented in Figs 6 and 7 which shows the relative percentage of AOM, phytoflagellates and palynomorphs (both marine and terrestrial) in each sample as single points in Tyson's Ternary Diagram (Tyson, 1995; Miles, 2021; Supplementary Files 3 and 4). The dinoflagellate cyst contribution in the carbonates is generally low, varying between 0.3% and 3.2%. This is also the case with cuticles (0.3–2.9%) and AOM (0.9–4.2%). For spores the general trend lies between ~10% and ~20%, and for pollen grains it is ~3–15%. Both spores and pollen grains have a similar outlier in Seep 8, where they comprise only 1.0% of the total counts. The Seep 8 sample also differs from the other carbonate samples in containing 47.7% coaly particles counting in the total assemblage. In the remaining carbonate samples coaly particles comprise between ~18% and ~33%. In the Seep 4 sample, the plant debris constitutes 51.0% of the palynomorph assemblage. For the other carbonate samples this value is between ~15% and ~27%. Woody particles in the carbonate samples vary between ~15–30%, apart from a lower outlier in Seep 4 with only 9.4%.

In the mudstone samples (85.8–87.6 m interval) below Seep 4, spores contribute between ~6% and ~18%, pollen grains ~5–10% with a higher value at level 86.7 m with 18.4%. Dinoflagellate cysts contribute between ~1% and ~3%, with an outlier at level 86.3 m with 4.5%. The proportion of cuticles is ~1.7% in these samples, with a higher contribution at level 86.3 m, where they contribute 4.2%. Most of the samples have a larger portion of woody particles than coaly particles, except for the sample level 86.9 m, where the coaly material counts for 26.1% and woody particles only for 21.7%. The palynomorph group that makes up the most of these mudstone samples is plant debris, varying between ~25% and ~35%. AOM is generally equal in the samples, ~3.7%.

At sample level 86.9 m, the palynomorph groups that dominate the palynofacies assemblage are coal (20.8–25.9%), woody particles (20.8–23.3%) and plant debris (30.6–33.7%). AOM, cuticles and dinoflagellate cysts each make up less than 5% of the total assemblage.

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3 Supplementary File 3: Palynology similarity indices; Supplementary File 4: Palynofacies organic particle data
Spore contents vary between 10.6% at 103.3 m and 9.1% at 104.8 m. In the upper of the two mudstone samples in our zone 3 (at 116.3 m), coaly particles make up the largest part with 37.6%, while plant debris and woody particles make up 27.4% and 20.2% respectively. The same groups dominate in the lower sample from zone 3 (at 115.0 m), but in this plant debris makes up the largest fraction with 24.9%. Woody and coaly particles both make up ~20% of this sample. The dinoflagellate cyst fraction is very low in the zone 3 samples, with 0.6% in the upper sample, while in the lower sample the value reaches 3.4%. AOM is close to the same value in both samples (4.4% and 4.8%) and cuticles in the lower sample constitute 4.0% while being only 1.9% in the upper sample. Spores and pollen make up a larger part in the lower sample, with 9.3% and 12.8% respectively, than in the upper sample where they are 2.9% and 4.8%.

We use the results of the palynofacies analyses to interpret the possible depositional environment of the carbonate seeps and mudstones in our section. Palynofacies alone is not a definitive way to determine a depositional environment, but it is useful as a supplement to other paleontological and sedimentological observations (Batten and Stead, 2005). The carbonate samples and the mudstone samples all plot within palynofacies fields I and III (Fig. 6), which correspond to “highly proximal shelf or basin” and “heterolithic oxic shelf (proximal shelf)” of Tyson (1995) respectively. The carbonate samples have a slightly greater spread in the diagram. With respect to kerogen type, the palynofacies field I represents kerogen type III which is gas prone, and palynofacies field III represents kerogen type III or IV, both are gas prone kerogen. Based on the position of the samples in the ternary diagram and variations in terrestrial vs. marine palynomorphs, we interpret that the seep containing sedimentary sequence at Kuhnpasset was deposited in a proximal shelf environment, shallower than the majority of the Stratumbjerg Formation in Northeast Greenland, which Bjerager et al. (2020) suggested was predominantly deposited in a basin-floor and slope environment.

CONCLUSIONS

From the palynostratigraphic and palynofacies analyses performed on six hydrocarbon seep carbonates, one carbonate nodule and 13 associated mudstone samples from the Kuhnpasset Beds, Wollaston Forland, Northeast Greenland, we draw the following conclusions:

– We propose three informal dinoflagellate cyst zones from the samples. Based on
stratigraphically important species, our lower zone 1 is assigned an ?early–late Barremian age, our middle zone 2 is late Barremian and our upper zone 3 is early Aptian in age.

- Based on dinoflagellate cyst biostratigraphy, hydrocarbon seepage in the Kuhnpasset area spanned the early – late Barremian age range, but did not continue into the Aptian. This indicates seep activity continued for as much as three million years, although the majority of seepage would have occurred over a shorter period of time, perhaps confined to the late Barremian.

- Statistical analysis shows a generally high dinoflagellate cyst diversity with a slightly lower diversity in the carbonate samples compared to the mudstone samples. The two uppermost mudstone samples differ from the rest of the studied material by yielding the lowest diversity and poorest preservation of dinoflagellate cysts.

- The dinoflagellate cyst assemblages recorded in this study show close similarities with other assemblages of similar age recorded in the Arctic/Boreal region.

- Based on the observed palynofacies material and relative proportions of marine and terrestrial palynomorphs, the carbonates and mudstones analyzed in this study were deposited in a proximal shelf setting.

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