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Characterization of microbial activity in pockmark fields of the SW-Barents Sea

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Abstract

Multibeam bathymetry revealed the occurrence of numerous craterlike depressions, so-called pockmarks, on the sea floor of the Hammerfest Basin and the Loppa High, south-western Barents Sea. To investigate whether these pockmarks are related to ongoing gas seepage, microbial processes associated with methane metabolism were analyzed using geochemical, biogeochemical and microbiological techniques. Gravity cores were collected along transects crossing individual pockmarks, allowing a direct comparison between different locations inside (assumed activity center), on the rim, and outside of a pockmark (reference sites). Concentrations of hydrocarbons in the sediment, particularly methane, were measured as headspace (free) gas, and in the occluded and adsorbed gas fraction. Down to a depth of 2.6 m below sea floor

(mbsf) sulfate reduction rates were quantified by radiotracer incubations. Concentrations of dissolved sulfate in the porewater were determined as well. Neither the sulfate profiles nor the gas measurements show any evidence of microbial activity or active fluid venting. Methane concentrations and sulfate reduction rates were extremely low or even below the detection limit. The results show that the observed sediment structures are most likely paleo-pockmarks, their formation probably occurred during the last deglaciation.

1 Introduction

Pockmarks are craterlike depressions in the seabed that form due to expulsion of fluids (Hovland and Judd, 1988). Fluids can be aqueous liquids like porewater (Harrington, 1985; Whiticar, 2002) but many published examples indicate formation by seeping hydrocarbons, especially methane, which can be of biogenic (Vaular et al., 2010) or thermogenic origin (Gay et al., 2006; Solheim and Elverhøi, 1985).

Pockmarks were first discovered in the late 1960's in Nova Scotia (King and MacLean, 1970), since then they have been observed globally in many different geologic settings, e.g. lakes, continental slopes, shelves and rises, shallow bays, fjords, as well as in the deep sea (Hovland and Judd, 1988; Judd and Hovland, 2007). They occur in different shapes e.g. as circular craters or as non-circular, elongated forms (Hovland et al., 2002). Sizes generally range from 10 to 200 m with depths up to 35 m, but also giant pockmarks with diameters of up to 1 km or more have been reported (Cole et al., 2000; Ondréas et al., 2005). Pockmarks can appear as single features (Prior et al., 1989) or in aggregations of hundreds of pockmarks, extending over tens of square kilometers (Lammers et al., 1995).

Hovland and Judd (1988) proposed a general model for pockmark evolution. Gas from greater depth migrates upwards through the sediment towards the seabed where it accumulates and causes doming. Stretching of the seabed causes small tensional fractures. Gas migrating along routes towards those fractures establishes a hydraulic connection, which results in a pressure drop followed by a violent burst. A unit pockmark is formed. Clusters of unit pockmarks can coalesce to a normal pockmark. A model by Woolsey et al. (1975) simulating “diatreme emplacements by fluidization” supports this theory. Recently, Cathles et al. (2010) came up with a conceptual model of pockmark and gas chimney formation. They assume that there is a capillary seal which traps the gas in the deeper sediment. Gas accumulation leads to overpressure and seal failure and an upward migrating gas diapir forms. With gas approaching the seabed, the sediment becomes fluidized and is removed, pockmarks begin to develop.

Pockmark formation is often associated with tectonic or sedimentary features such as faults or buried channels (Chand et al., 2008). It can be controlled by underlying bedrock and lithological boundaries (Forwick et al., 2009), but can also be triggered by extreme events like earthquakes and tsunamis, melting of grounded icebergs or even by human influence (Hovland et al., 2002).

There are only few examples in which active fluid or gas flux could be observed. However, active pockmarks have been found all over the world (Gay et al., 2007; Lammers et al., 1995; Ondréas et al., 2005; Solheim and Elverhøi, 1985) but also inactive, dormant pockmarks (Hovland and Judd, 1988; Ussler et al., 2003; Webb et al., 2009), even entire inactive pockmark fields (Paull et al., 2002; Solheim and Elverhøi, 1993) and buried paleo-pockmarks, discovered by 3D seismic data (Cole et al., 2000; Hartwig et al., 2012, this issue), have been described.

Leakage of hydrocarbons through pockmarks infers that there is a gas source in the underlying sediment. As potential indicators for deeper hydrocarbon reservoirs, pockmarks received considerable interest from the hydrocarbon exploration industry (Hasiotis et al., 2002; Hovland and Judd, 1988).

Submarine hydrocarbon seeps, such as pockmarks or mud volcanoes, are habitats for specific microbial communities. Epifaunal and bacterial aggregations like thiotrophic bacterial mats associated with these structures are widely described in the literature (Foucher et al., 2009; Niemann et al., 2006) and provide evidence of ongoing nutrient provision and metabolization. Pockmarks also play an important role in the global methane cycle. Methane coming from a source at greater depth can be released to the overlying ocean and even partly to the atmosphere (Lammers et al., 1995; O'Connor et al., 2010; Reeburgh, 2007), effectively bypassing the zone of anaerobic oxidation of methane, which normally prevents any methane escape from sediments.

In subsurface sediments microbial activity dominates, with dissimilatory sulfate reduction as the quantitatively most important electron acceptor process in the anaerobic degradation of organic matter (D'Hondt et al., 2002; Jørgensen, 1982). Thus, determination of sulfate reduction rates provides a reasonably good estimate of total microbial activity in anoxic marine sediments. According to the carbon source used, sulfate reduction (SR) can be divided into two main pathways.

Organiclastic sulfate reduction takes place in the upper part of the sediment close to the sediment-water interface. It is fuelled by small organic molecules like fatty acids, derived from the degradation of organic matter. The organic matter is oxidized by dissolved sulfate, which is in turn reduced to hydrogen sulfide (Jørgensen, 1982; Martens and Berner, 1974), according to equation 1.

Eq. 1 $2\ CH_2O + SO_4^{2-} \rightarrow 2\ HCO_3^- + H_2S$

Methanotrophic SR, also known as anaerobic oxidation of methane (AOM), occurs below the zone of organiclastic sulfate reduction in a narrow band where there is an overlap of dissolved sulfate and methane (Martens & Berner 1974, Iversen & Jørgensen 1985), the so-called sulfate-methane transition zone (SMTZ). The reduction of sulfate to sulfide is facilitated by the oxidation of methane to bicarbonate (equation 2).

Eq. 2 $CH_4 + SO_4^{2-} \rightarrow HCO_3^- + HS^- + H_2O$

A consortium of archaea and sulfate-reducing bacteria (SRB) has been reported to facilitate the anaerobic oxidation of methane with sulfate as an electron acceptor (Boetius et al., 2000; Orphan et al., 2001). Recently, other electron acceptors have been identified, e.g. Nitrate (Raghoebarsing et al., 2006).

Below the SMTZ sulfate is usually not detectable in the porewater, some very minor concentrations (single to tens of µM) are usually attributed to reoxidation of hydrogen sulfide during sample processing. However, molecular studies could identify sulfate reducing microbes below the SMTZ (Schippers et al., 2010), even in the zone of active methanogenesis (Leloup et al., 2007).

In this study, three areas in the southwestern Barents Sea were investigated (Fig. 1). Two of these areas (areas B and F) are part of a large field of many small depressions in the seabed (Fig. 2), which were identified as pockmarks. The third

area (area D) does not contain any pockmark features but was chosen for further investigations due to an assumed underlying gas anomaly, as interpreted from seismic data. Since pockmarks could be valuable indicators for deeper hydrocarbon reservoirs, we investigated whether these pockmarks are related to ongoing gas seepage or if they are manifestations of a past event. About 200 sediment gravity cores were collected and analyzed. A combined study of gas measurements, quantification of geochemical parameters and microbial activity was conducted to obtain a better understanding about ongoing processes in and around the pockmarks and to reveal their formation history.

2 Geological setting

The Barents Sea is a deep epicontinental sea, which borders to the Norwegian Sea in the southwest and to the Arctic Ocean in the north. It is characterized by several shallow banks and is crossed by deep troughs and faults (Fig. 1). The investigated area is located between the southwestern part of the Loppa High, comprising the Polheim sub-platform, and the Ringvassoy-Loppa Fault complex and defined by a square with SW and NE coordinates 72°13'N, 20°38'E and 71°57'N, 19°53'E (Fig. 1).

The Loppa High is a structural high near the south-western margin of the Norwegian Barents Sea. Fault complexes separate the Loppa High from the Hammerfest basin in the southern and the Bjørnøya Basin in the western part. In the south-east it passes into the Hammerfest Basin and the Bjarmeland Platform, separated by a monocline.

The area has undergone a complex geological history, characterized by several phases of uplift/subsidence and subsequent tilting and erosion (Larssen et al., 2005), whereby the Loppa High is a result of Late Jurassic to Early Cretaceous tectonism

(Gabrielsen et al., 1990). According to Eidvin et al. (1993) and Cavanagh et al. (2006) the latest erosion phase is linked to the glacial period.

During the Last Glacial Maximum (LGM) the study area was totally covered by grounded ice (Andreassen et al., 2008; Siegert et al., 2001). During that time the ice sheets reached the shelf break twice (LGM I and LGM II) in the SW-Barents Sea. LGM I occurred at 19 ka followed by the Andoya Interstadial which commenced with a high arctic climate after which middle to low arctic climate prevailed. Large areas of the southern Barents Sea were deglaciated again. At 18 ka the LGM II occurred in this area (Junttila et al., 2010; Vorren and Laberg, 1996). Between 16 and 15 ka a warm spell caused the thinning of the ice sheets, triggering a major deglaciation. A major drawdown of the marine-based Barents Sea Ice Sheet occurred around 14.5 ka. As of roughly 12 ka the study area was totally free of ice.

3 Materials and Methods

3.1 Acoustic survey and Sampling

In 2009 the Forsvarets Forskenings Institutt (FFI) conducted a multibeam bathymetric survey of the Hammerfest/Loppa High and the Tromsø Basin/Ingøydjupet area of the western Barents Sea region, using an EM 710 echo sounder system, with a medium frequency of 70-100 kHz. During a cruise in November 2009 on R/V HU Sverdrup to the Loppa High about 200 sediment gravity cores with a maximum length of 2.6 m were taken in three different areas (B, D, F).Topographic seafloor maps of the three investigated areas (B, D, F) showing the exact sample locations are given in

Figure 2. To map the general gas distribution within each area, between 15 and 115 cores were taken for gas analysis. Due to the massive amount of cores for gas analysis, samples were only taken from the deepest part of each core. Two sediment samples of ca. 500 cm^3 each were taken and immediately transferred into gas-tight metal cans and stored at -80°C until analysis after the cruise.

A total of 35 cores (exact locations are given in Table 1) were selected for detailed microbiological and geochemical studies. Each of these cores was sampled in ten depth intervals directly on board. To avoid contamination from shallower sediment being dragged down during penetration of the core, the exterior of the material was shaved off prior to subsampling.

Sampling for SRR incubation experiments was carried out by inserting 5-mL tip-cut plastic syringes into the sediment, thereby retrieving small subcores with an intact sedimentary structure. After retrieval of the subcore, the open end was closed with a butyl rubber stopper and the syringes stored in nitrogen-flushed gas-tight bags in a refrigerator close to in-situ temperature (+5°C). Usually this type of incubation is carried out in gas-tight glass barrels. Unfortunately our glass barrels got lost during transport, so we had to revert to plastic syringes. Although plastic syringes are less than perfect for incubating SRR samples because of their permeability for gasses, in this case the potential bias should be minimal because there is basically no methane in the sample which could potentially be lost. Changes in pH due to loss of CO_2 can also be neglected because the porewater is well-buffered with regard to its carbonate system. An aliquot of the sediment was frozen at -20°C in N_2-flushed gas-tight bags for pore-water extraction after the cruise.

3.2 Sulfate reduction rates (SRR)

Sulfate reduction rates were determined using a modification of the whole core $^{35}SO_4^{2-}$ injection method (Jørgensen, 1978) as described by Ferdelman et al. (1999) and Treude et al. (2005b). All incubations were performed in duplicates.

4 µl of carrier-free $^{35}SO_4^{2-}$ (180 kBq/µl) were injected along the longitudinal axis of the subcore. The incubations were conducted for 24 hours and terminated by transferring the radiolabeled sediment into 10 mL of 20% (w/v) zinc acetate (ZnAc). Blank samples were prepared by injecting $^{35}SO_4^{2-}$ radiotracer into a sample, followed by immediate transfer into 20% (w/v) zinc acetate. Additionally, tracer blanks were prepared by adding 4 µl of radiotracer directly into ZnAc without any sediment. The total reduced inorganic sulfur (TRIS) compounds (mono- and disulfides, elemental sulfur) were separated using the cold chromium distillation method (Kallmeyer et al., 2004). Radioactivity was quantified using a Perkin Elmer Tri Carb 2800 TR liquid scintillation analyzer and Perkin Elmer Ultima Gold XR Scintillation cocktail. SRRs were calculated using the following equation (Jørgensen, 1978; Kallmeyer et al., 2004):

$$SRR = \frac{a_{TRIS}}{a_{TOT}} \times \frac{[SO_4^{2-}]}{t} \times \rho_{SED} \times 1.06 \times 1000000$$

where a_{TRIS} and a_{TOT} are the activities of the TRIS compounds and the total activity of the injected $^{35}SO_4^{2-}$, respectively, using the raw counts (given as counts per minute, cpm) from the scintillation counter; t is the incubation time and ρ_{SED} the porosity of the sediment (mL porewater cm^{-3} sediment). SO_4^{2-} is seawater SO_4^{2-}

concentration, which was set to 28 mM for all samples, because there was no measurable down-core depletion of porewater SO_4^{2-} concentrations in any of our samples. 1.06 is a correction factor for the expected isotopic fractionation (Jørgensen and Fenchel, 1974), 1000000 is the factor to change units from mmol L^{-1} to pmol cm^{-3}.

3.3 Determination of gas concentrations

GeolabNor (Trondheim, Norway) quantified C_1-C_6 hydrocarbons in the headspace (free) and occluded gas fraction as well as C_1-C_8 hydrocarbons in the adsorbed gas fraction, using gas chromatography. Headspace gas was determined by thawing the frozen samples over night with subsequent analysis of the free gas in the headspace of the container. Occluded gas was released mechanically from an aliquot of the sediment using a ball mill under vigorous shaking. Treatment of an aliquot of sediment with orthophosphoric acid led to the detachment of the gas adsorbed to clay minerals. All values are shown in the unit µmol L^{-1} sediment (µM). Extrapolated gas distribution maps were generated based on the obtained gas data.

3.4 Porewater sulfate concentration

Sediment porewater was obtained by centrifugation and careful decantation followed by filtration through a 0.2 µm membrane. Porewater sulfate concentrations were determined by suppressed ion chromatography using a SYKAM IC-System (injection volume 50µL, anion exchange column, SYKAM LCA A14; conductivity detector, SYKAM S3115).The elutant was a 6.25 mM sodium carbonate solution with 0.1% (v/v) modifier (1 mL 4-hydroxy-benzonitrile in 50 mL methanol), the flow rate was 1mL min^{-1}.

4 Results

4.1 Morphology of seabed

The morphology of the seabed in our study areas is shown in Figure 2. Multibeam bathymetry revealed the presence of a multitude of circular seafloor depressions interpreted as pockmarks. Area B and F contain an average density of 100 pockmarks per square kilometer, either randomly distributed or in arrays along iceberg plough marks, which are the result of former iceberg activity, formed primarily during glacial retreat (Solheim, 1997). The up to 15 m deep plough marks can be observed all over the seabed surface.

The circular pockmarks are between 10 and 50 m in diameter with an average depth of about 1-3 m. According to Hovland et al. (2002; 1988) these pockmarks can be termed “normal pockmarks”. Additionally, some larger, irregular pockmarks with diameters of up to 300 m and depths up to 25 m were observed. As mentioned before, area D does not contain any pockmark features, but, like in area B and F, many plough marks could be seen on the seabed of area D.

4.2 Samples

The average water depth in the investigated area was found to be around 350 m with an average water temperature of about 6°C. Sediment composition appeared to be very similar in all three areas, being brown to dark brown in color. Except for the first 10 cm which were softer, porosity averaged around 0.52 throughout the entire length of the core.

In general the cores consisted of very sticky silty clay, occasionally containing some less dense structures like small pieces of siliceous sponges as well as pebbles or fragments of bivalve shells. There was no difference between sediment from within or

from outside the pockmarks. No hydrogen sulfide smell was noticed in any core nor were there any obvious authigenic carbonate structures.

In area D the penetration depth of the gravity corer was relatively small (average penetration depth of 50 cm). The sediment material above the anomaly structure consisted of very sticky mud preventing a deeper penetration.

4.3 Gas concentrations

Concentrations of hydrocarbons (methane and higher n-alkanes, cyclic and branched hydrocarbons) in the headspace (free), occluded, and adsorbed gas fraction were determined on samples from the deepest interval of the core (maximum depth 2.6 mbsf). Extrapolated gas distribution maps were created to show the distribution of hydrocarbons in the three study areas. All maps show extremely low concentrations and do not reveal any correlation between gas concentration and sampling sites (pockmark or reference site). The following discussion is focused just on adsorbed methane, since this fraction exhibited highest concentrations. In the occluded and free gas fraction only marginal amounts could be found, concentrations of longer chain hydrocarbons were also negligibly low.

Table 2 summarizes some statistic values for the adsorbed methane and free methane fraction of the three study areas. Figure 3 shows exemplarily the distribution of adsorbed methane in area B. In area B the highest methane concentration in the adsorbed gas fraction was 29.1 µM. Average values were only 6.19 µM in the pockmark cores and 5.93 µM in reference cores (Tab. 2, Fig. 3).

In area D the maximum concentration of adsorbed methane was 16.84 µM, with an average value of 5.42 µM. In area F concentrations of adsorbed methane reached a

maximum value of 19.18 µM, the average values for the pockmark samples 6.63 µM and for the reference samples 5.78 µM. There are some local maxima in gas concentration in all three areas, but even these anomalies are still very low and they do not generally correlate to pockmarks.

Due to the very low gas concentrations no isotope measurements could be carried out. Consequently it remains unknown whether the gas is coming from a biogenic or thermogenic source.

4.4 Sulfate-profiles

On 12 cores porewater sulfate concentrations were determined in 10 depth intervals between 0-240 cm. The profiles show very little variation with depth and among each other (Fig. 4). The almost linear profiles and the only minimal decrease with depth indicate very low net sulfate consumption. When extrapolating linearly, the calculated SO_4^{2-} penetration depth is 37 meters, a value that is more common for oligotrophic open ocean sites, not for ocean margins. For gas rich sediments, this value normally ranges between 0.02 – 0.2 m (Luff and Wallmann, 2003; Treude et al., 2005a).

4.5 Sulfate reduction rate

Exemplary SRR depth profiles for pockmark and reference sites in area B are presented in Figures 5a to 5d. Irrespective of pockmark or reference site, most of the sulfate reduction activity was restricted to the upper 30 cm of the sediment (Figs. 5a and b). In this depth interval the highest observed SRR was 600 pmol/cc/d. Deeper than 30 cm below the sea floor (cmbsf) measurements fell mostly below the minimum

detection limit (MDL) of 10 pmol cm^{-3} d^{-1} (Fig. 5a). Only a few single samples showed higher values of up to 200 pmol cm^{-3} d^{-1} (Fig. 5b), but even those maximum values are still very low when compared with other sites from literature e.g. active seeps (Boetius et al., 2000; Pimenov et al., 2010; Treude et al., 2003). Even in the permanently cold sediments of Svalbard in the northern part of the Barents Sea, sulfate reduction rates are much higher (around 60 nmol cm^{-3} d^{-1}) (Finke et al., 2007). In some cores all values fell below the MDL, irrespective whether the site was located within or outside of a pockmark (Figs. 5c and d).

In area F (Figs. 5e and 5h), highest SRR was also found in the top layer. For the majority of the samples, sulfate reducing activity was restricted to the upper 20 cm of the sediment, reaching a maximum of up to 400 pmol cm^{-3} d^{-1} (Fig. 5f). Below this depth interval most values were close or below the MDL (Fig 5g and 5h), slightly higher values could only be detected in very few samples. In several cores from both pockmarks and reference sites, SRR fell continuously below the MDL (Fig. 5h).

In area D, SRR measurements fell consistently below the minimum detection limit in all samples.

In general, sulfate reduction rates do not show any difference between cores from within or outside of pockmark structures. Profiles with slightly higher values in the upper part as well as profiles showing all values below the MDL have been detected for both pockmark and reference sites. Furthermore all SRR profiles show values close to the detection limit, indicating a generally very low microbial activity in the sediments of the investigated area.

5 Discussion

A multi-proxy approach was undertaken to obtain chemical, geomicrobiological and geochemical evidence for active fluid or gas venting. Seafloor seeps that contain methane or hydrogen sulfide can easily be identified by their sediment porewater chemistry, their association with specific microbial communities and high rates of microbial turnover as well as characteristic features in the seismic data. However, in our study area there was no evidence for active fluid venting currently occurring from either type of analysis.

During sampling we did not notice any hydrogen sulfide smell, not even on acidified porewater samples, implying that there are neither significant amounts of dissolved H_2S nor any finely precipitated sulfides such as ferrous monosulfide (FeS). Furthermore, no apparent carbonate concretions, being an indicator for active or recent anaerobic oxidation of methane, could be identified (Ferrell and Aharon, 1994; Knittel and Boetius, 2009).

5.1 Gas distribution maps

Gas concentrations in sediments of the investigated areas were extremely low. In some single samples, slightly elevated gas concentrations could be detected but no consistent correlation with the occurrence of pockmarks or any other parameter could be established. Headspace gas and occluded gas concentrations were negligibly small, implying that no active seepage is taking place in the pockmark structures. Highest values were always detected in the adsorbed gas fraction, it is therefore reasonable to assume that this gas is derived from previous gas emissions, during which a portion of the gas was adsorbed onto the clay minerals.

There are no significant differences between the concentrations of adsorbed methane in all three areas. Throughout our study site, the average gas content is

very low and shows no correlation between sites from within or outside the pockmarks. Very low gas contents in area D demonstrate that there is no connection between the potentially underlying gas anomaly and the surface sediments. Due to the fact that gas concentrations were always below saturation and no bubble formation occurred, any major loss due to degassing during sampling and processing can be ruled out.

5.2 Sulfate reduction rate (SRR) & Sulfate-profiles

Unlike methane, which can be lost from the sediment during coring due to degassing, porewater sulfate cannot be lost as it does not form a gaseous phase. In sulfidic sediments however, there is the potential for increasing sulfate concentrations due to oxidation of hydrogen sulfide. Because of the lack of hydrogen sulfide, we can rule out this possibility and assume the sulfate profile to represent true in-situ conditions.

The linear decrease in porewater sulfate concentration at all sites indicates that diffusion dominates sulfate transport into the sediment and that there is no sulfate consumption in this depth interval. As described by Berner (1980) SR activity can be estimated from porewater sulfate profiles. However, direct radiotracer measurements of SRR provide much more reliable results of SR activity, especially if diffusion is not the only means of transport and other processes like advection may replenish the sulfate pool in near-surface sediments (Fossing et al., 2000; Jørgensen et al., 2001; Weber et al., 2001). This is the case in our study because contrary to the linear sulfate profiles, which indicate no net sulfate consumption over the length of the core, direct radiotracer SRR measurements reveal some very low activity in the upper few cm. Much higher rates of sulfate reduction were expected in the pockmark cores, particularly if active fluid venting was taking place. In most cases SR was only

389 measurable in the upper 30-40 cm below sea floor (cmbsf). This also leads to the
390 assumption, that AOM does not occur in the recovered depth interval (2.6 m) and the
391 SR-activity can be attributed entirely to organiclastic SR and not to anaerobic
392 oxidation of methane.

393 The calculated SO_4^{2-} penetration depth of 37 meter is quite deep when compared to
394 data of gas-rich sediments. In Eckernförde Bay (German Baltic Sea) for example,
395 Treude et al. (2005a) determined a penetration depth of only 0.32 m. Such a deep
396 sulfate penetration depth shows that very little sulfate is consumed in the upper part
397 of the seabed. Sulfate reduction rates in our study area are generally very low, with
398 no significant differences between SRR profiles from pockmark cores and reference
399 cores. Additionally, SRR values in area D fall consistently below the minimum
400 detection limit in all samples. This leads to the assumption that the pockmarks do not
401 release any gas at present, which is supported by our findings of very small
402 decreases in porewater sulfate concentration with sediment depth and very low gas
403 concentrations.

404

405 **5.3 Pockmark formation theory**

406 Fluid and gas venting is commonly believed to be the main factor for pockmark
407 generation. While pockmark structures could clearly be detected on the seabed, we
408 did not find any indications for significant concentrations of methane in the sediments
409 of the sampling area. Neither did we find indications for ongoing fluid flow through
410 these features.

411 Although expulsion of methane is the most frequently reported process leading to the
412 formation of pockmarks, seepage of other fluids has been described as well.
413 However, in most cases only minor quantities of gases like hydrogen sulfide or

carbon dioxide are associated with hydrocarbon leakage. The expulsion of porewater through the seabed, so called submarine groundwater discharge (SGD), has been widely described in the literature. In a review Taniguchi et al. (2002) describe different systems showing SGD.

Some examples of pockmarks being formed by porewater discharge, have been reported (Harrington, 1985; Webb et al., 2009; Whiticar and Werner, 1981). However, those few examples are primarily based on assumptions and lacking direct evidence of the processes involved. Moreover, the mechanisms described in these studies are unable to explain the procedure of pockmark formation in detail. Given the enormous area of extensive pockmark occurrence within and beyond our study area and the fact that we have no evidence for past or present SGD, we rule out this mechanism.

Recently the occurrence of unit pockmarks (pockmarks diameter < 5 m) was discussed to be an indicator for active gas seepage (Cathles et al., 2010; Hovland et al., 2010).

Mapping unit pockmarks requires application of high-resolution bathymetry (at least 1m x 1m gridding) (Hovland et al., 2010). The resolution of the bathymetry equipment which was used in this study is too low to locate unit pockmarks. However, due to the very low gas concentrations in the pockmark as well as in the reference sites (table 2) we rule out the existence of seeping hydrocarbons in our study area.

Pockmark structures which are present in the seabed consistently over several years are not necessarily an indicator for active fluid flow (Brothers et al., 2011). Numerical modeling by Hammer et al. (2009) showed that upwelling currents can be a possible mechanism to maintain pockmark structures even if activity has ceased for several years.

This leads us to the more likely hypothesis that the pockmarks in our study area are preserved features from past activity. The present seabed shows many plough marks, derived from iceberg drifting during the last deglaciation. As undisturbed pockmark craters can be found within and outside of the plough mark structures, the pockmarks must have formed during and after the main phase of iceberg movement.

Considering the glacial history of the Barents Sea as well as the great number of glacial plough marks in the study area, it can be assumed that the whole area was covered by a grounded ice sheet during Last Glacial Maximum (ca. 20 ka ago). Gaseous hydrocarbons like methane, derived from a deeper lying source, could have migrated through the porous sediment towards near-seabed sediments. Due to the conditions during the glacial period, including pressure of a thick grounded ice sheet affecting the sediment, combined with the prevailing low temperatures, the necessary conditions for clathrate formation were given (Kvenvolden, 1998; Sloan, 2003). Methane could thus have been trapped as gas hydrate in the shallow sediment.

The uplifted Atlantic margin basins, comprising the Hammerfest Basin and the Loppa High, contain several oil and gas fields commonly occurring in underfilled traps. Corcoran and Doré (2002) suggested that this was caused due to large scale leakage of gas during the Cenozoic exhumation. 2D basin modeling of this area by Cavanagh et al. (2006) revealed that glacially controlled pressure oscillations provide a mechanism for episodic discharge of methane from the deep petroleum reservoirs and sequestration of methane as gas hydrates near the surface.

Alternatively sub-glacial gas hydrates may have been fed by a biogenic source or a combination of biogenic and thermogenic gases.

About 14 ka ago the climatic conditions of the SW Barents Sea changed within a relatively short time interval (approx. 5000 years, Elverhøi *et al.*, 1995), causing deglaciation, melting of the ice sheet and iceberg calving. Thus, the conditions to preserve hydrate layers no longer persisted, resulting in the rapid decomposition of the gas hydrate as well as the release of potentially accumulated gas. Such an event probably occurred over large areas of the Barents Sea more or less simultaneously, resulting in widespread pockmark structures on the seabed. Similar formation mechanisms have already been described for other areas (Davy et al., 2010; Long, 1992; Long et al., 1998; Mienert et al., 1998; Plaza-Faverola et al., 2012; Solheim and Elverhøi, 1993; Sultan et al., 2010).

Maslin et al. (2004) collected published data from submarine sediment failures and correlated them with climatic changes from the past 45 ka. They found that over 70% of continental slope failures could be dated to the periods between 15-13 ka and 11-8 ka. These dates, as well as the rising sea level and global atmospheric methane records, correlate well with the timing of the last deglaciation. The hypothesis of pockmark formation due to rapid methane release caused by destabilization of gas hydrates during this time is supported by ice-core data (Chappellaz et al., 1993).

From the above discussion we can summarize the following points concerning the proposed scenario on the formation of the Loppa High pockmarks:

1. During LGM a grounded ice sheet existed in the study area. Thermogenic and / or biogenic gas from a source in the Mesozoic bedrock migrated through the sediment and accumulated as gas hydrate, which was stable under the given pressure and temperature conditions(Fig. 6 a).

2. The retreat of the ice sheet during deglaciation caused changes of pressure conditions, which resulted in the decomposition of the hydrate layer and the release of large volumes of gas over a large areal extent, resulting in the formation of seafloor pockmarks (Fig. 6 b).

3. Nowadays the pockmark structures are still preserved despite the seeping activity having ceased several thousand years ago (Fig. 6 c).

The depressions seem to be relatively young features. As many pockmarks are located inside the iceberg plow marks, it can be assumed that the pockmarks were formed postglacially, probably in an early phase after the last deglaciation. Similar observations have also been reported from the northern Barents Sea (Solheim and Elverhøi, 1993) and could be an indication for a large methane release event caused by the decay of the Barents ice sheet during the last deglaciation.

6 Conclusions

Deep, craterlike structures, identified as pockmarks were observed by multibeam bathymetry on the seafloor of the SW-Loppa High in the SW-Barents Sea. Geochemical, biogeochemical and microbiological analysis revealed

1. Very low concentrations of hydrocarbons in the sediment.

2. Almost no depletion of porewater sulfate with depth.

3. Low sulfate reduction rates, close to detection limit.

4. Currently no anaerobic oxidation of methane in upper 3 mbsf.

5. No significant and consistent differences in any measured parameter between sites inside and outside of pockmarks.

This indicates astonishingly low microbial activity in the study area, which supports the assumption that currently no active fluid venting is taking place. Based on our own findings and the current literature we hypothesize that the pockmark occurrence and its formation is most likely related to a paleo-event during the last deglaciation.

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