1	Hydrothermal vent complexes offshore Northeast Greenland: a potential role in driving
2	the PETM
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16	ABSTRACT
17	Continental rifting is often associated with voluminous magmatism and perturbations in the
18	Earth's climate. In this study, we use 2D seismic data from the northeast Greenland margin to
19	document two Paleogene-aged sill complexes ≥18 000 and ≥10 000 km² in size. Intrusion of
20	the sills resulted in the contact metamorphism of carbon-rich shales, producing thermogenic
21	methane which was released via 52 newly discovered hydrothermal vent complexes, some of
22	which reach up to 11 km in diameter. Mass balance calculations indicate that the volume of
23	methane produced by these intrusive complexes is comparable to that required to have caused
24	the negative $\delta^{13}C$ isotope excursion associated with the PETM. Combined with data from the

conjugate Norwegian margin, our study provides evidence for margin-scale, volcanically-

induced greenhouse gas release during the late Paleocene/early Eocene. Given the abundance of similar-aged sill complexes in Upper Paleozoic-Mesozoic and Cretaceous-Tertiary basins elsewhere along the northeast Atlantic continental margin, our findings support a major role for volcanism in driving global climate change.

Keywords

Hydrothermal vent complexes, sill intrusions, PETM, global climate change, Greenland, NAIP

Volcanic rifted margins are associated with voluminous extrusive and intrusive igneous

1. Introduction

activity (Menzies et al., 2002; Jerram and Widdowson, 2005) of which the northeast Atlantic margins are type examples (e.g. Saunders et al., 1997). Here, extrusive activity during the Paleocene and Eocene produced characteristic Seaward Dipping Reflectors and extensive subaerial lava flows, whilst intrusive activity produced igneous centres (Jerram and Bryan, 2015) and sill complexes ≥80 000 km² in size (Planke et al., 2005; Schofield et al. 2015).

The emplacement of sill complexes can generate huge quantities of greenhouse gases by metamorphic reactions within the intrusion aureole system (e.g. Aarnes et al., 2011, 2012, 2015). The composition and volumes of gases generated are dependent on a range of factors including (and most importantly) host rock composition, total organic content (TOC) and permeability; in addition to intrusion volume, temperature and emplacement depth (Aarnes et al., 2012; Iyer et al., 2013). A proportion of these gases are released to the atmosphere or hydrosphere through hydrothermal vent complexes within tens of years of sill intrusion (e.g. Jamtveit et al., 2004; Aarnes et al., 2010). Hydrothermal vent complexes commonly form above sill tip terminations as a result of intensive fracturing or brecciation of overburden strata in the shallow subsurface. These overburden breaches are caused by overpressure build up

associated with the boiling of pore fluids and host rock devolatilization reactions (Jamtveit et al., 2004; Aarnes et al., 2012). Where vent structures are observed in seismic data, they have eye, dome or crater-like upper parts (Planke et al., 2005; Møller Hansen, 2006). The lower parts are characterised by a central pipe, commonly surrounded by a region of inwardly dipping strata that is contained within metamorphosed sedimentary rocks (Møller Hansen, 2006; Svensen et al., 2007). Release of gases from these vents is thought to have played a primary role in driving global warming, as proposed for the Paleocene Eocene Thermal Maximum (PETM) (Svensen et al., 2004).

Extensive sill complexes are documented from the United Kingdom Continental Shelf (e.g. Schofield et al., 2015) and the Norwegian margin (Svensen et al., 2004) where hydrothermal vent complexes are also recognised. Whilst onshore studies have documented sills within Carboniferous—Cretaceous-aged sediments (Price et al., 1997; Therkelsen, 2016), sparse data coverage in regions covered by sea ice and poor seismic imaging beneath the "top basalt" reflection means that the offshore section of this margin has long been a significant gap in our understanding of the northeast Atlantic continental margins. Without the full extent of intrusive complexes in the North Atlantic Igneous Province (NAIP) being recognised and mapped, the role of margin-scale intrusive volcanic activity in driving global climate change has remained uncertain.

This study uses newly acquired 2D seismic data to document the distribution and architecture of sill complexes and associated hydrothermal vent complexes along the offshore northeast Greenland margin. A combination of seismic mapping, field evidence (e.g. Larsen and Marcussen, 1992) and burial history curves (Mathiesen et al., 2000) indicate that the sills within the Danmarkshavn Basin regionally intruded Jurassic-aged, shale-rich horizons at paleodepths of >3 km, whilst in the Thetis Basin they intruded Cretaceous-aged host rocks at paleodepths of 1–2 km. The Jurassic-aged shales have TOC (total organic carbon) contents up

to twenty times higher than those reported from the Norwegian margin (Svensen et al., 2004; Price and Whitham 1997). Contact metamorphism of these shales resulted in the voluminous production of greenhouse gases such as methane, released into to the atmosphere via hydrothermal vent complexes (Svensen et al., 2004; Aarnes et al., 2015). Combined with data from the Norwegian and United Kingdom Continental Shelf margins, we show that volcanically-induced greenhouse gases were produced on a scale capable of producing the observed negative δ^{13} C excursion during the PETM.

2. Dataset and Methods

This study utilizes 2D seismic profiles acquired by TGS in 2008–9 and 2011–14, including re-processed AWI data. The surveys cover an area of ~125 000 km², with the lines ranging from 40–250 km in length and spacings varying from 0.1–40 km. Seismic interpretation was conducted using Kingdom software. The extrusive and intrusive volcanic facies have been mapped on intersecting 2D seismic lines (Fig. 1) using the seismic volcanostratigraphic methods of Planke et al. (2000) and Planke et al. (2015). In the absence of well data we use a p-wave velocity of 5.5 km s⁻¹ to determine the resolution and detection limit for the sills (Skogly, 1998; Berndt et al., 2000) and a velocity of 1.8 km s⁻¹ to calculate the dimensions of the upper parts of the vents (Planke et al., 2005). The sedimentary basins are correlated with the onshore successions of east and northeastern Greenland, and offshore successions in the southern Barents Sea and on the mid-Norwegian shelf (Hamann et al., 2005; Tsikalas et al., 2005). More than one hundred gravity core and tens of dredges have been acquired to study hydrocarbon seepages and to allow seismostratigraphic ties.

3. Interpretation of sill and hydrothermal vent complexes

The sills are characterised by high amplitude, positive reflections, indicating a significant downwards increase in acoustic impedance. They commonly display abrupt terminations, saucer shaped morphologies and transgress the stratigraphy (Figs. 2 and 3); diagnostic features of igneous intrusions (e.g. Planke et al., 2005; 2015). The sills are dominantly found within two complexes; a \geq 18 000 km² complex in the Cretaceous-Tertiary age Thetis Basin and a \geq 10 000 km² complex in the Upper Paleozoic-Mesozoic-aged Danmarkshavn Basin. The sills within these complexes are also documented by Hamann et al., (2005) and Geissler et al., (2016). The sill complexes follow the structural trend of the basins and are oriented NNE/SSW (Fig. 2). Within the Thetis Basin the sills are up to 28 km in diameter and were emplaced at depths of 1–2 km; this is interpreted from their relationship to the Vent Horizon (see below). In the Danmarkshavn Basin the sills are up to 40 km in diameter and were emplaced at depths of 3–5 km. The sills in the Danmarkshavn Basin tend to be more layer parallel than those in the Thetis Basin, which are commonly saucer-shaped (Fig. 2). This morphological-depth relationship is typical of sill complexes (Planke et al., 2015).

Sills are not imaged beneath the extrusive facies (e.g. Inner Flows; see Planke et al., 2000) and are absent within the Danmarkshavn Ridge. Sparse data coverage prevents us from determining the northward extent of the complex in the Thetis Basin. The frequency of the seismic data at the depths at which the sills are found is 10 Hz, therefore the sills need to be >200 m thick to be resolved and >50 m thick to be detected. Although imaging of deep sills and distinguishing sills from multiples beneath the first high amplitude sill reflection event is challenging, intersecting seismic surveys indicate that the sills are vertically stacked, with each complex containing \geq 4 sills which decrease in number toward the basin margins.

Linked to the tips of the sills by vertical chimney zones of disturbed reflections are a series of hydrothermal vent complexes. These vents have eye-, dome- and crater-type upper parts, similar to those on the conjugate Norwegian margin (Planke et al., 2005). The eye- and dome-

type vents have sub-parallel, prograding internal reflections whilst the crater-type vents have internal reflections which vary from chaotic to parallel. We calculate the upper part of the vent complexes are 36–504 m in height. The diameter of the vents ranges from 0.7–11 km (e.g. Fig. 4). Within the Thetis Basin, the upper parts of all vents are located at a consistent stratigraphic horizon which is onlapped by overlying reflections; this is identified as the Vent Horizon (VH). Onlap relationships indicate the VH represents the paleosurface at the time of sill intrusion. Within the Danmarkshavn Basin, the VH forms the top of the crater-type vents. Towards the Volcanic Complex, the VH terminates against the Inner Flows (Fig. 5).

A total of 52 vent complexes have been identified from both the Danmarkshavn (n=17) and Thetis basins (n=35). However, it is likely there are many more hydrothermal vents which are not intersected by the available 2D seismic lines. The vents have previously been documented by Geissler et al., (2016) and are distinguished from superficially similar volcanoes by their stratigraphic position, lower amplitude and differing internal reflections (cf. Fig. 12 in Schofield et al., 2015 and Reynolds et al., 2016). The vents are distinguished from biogenic mounds since they are much smaller than these features and they do not form above faults (e.g. Langhi et al., 2016). Furthermore, the vents we describe are ubiquitously associated with sills (e.g. Figs. 3 and 6) typical of hydrothermal vents (e.g. Planke et al., 2005) and unlike other mounded structures (e.g. Hansen et al., 2005).

4. Determining the effects of thermogenic methane release

4.1 Host Rock Properties

The quantity of gas produced during contact metamorphism is greatly influenced by the host rock properties. In the Cretaceous-aged Thetis Basin, the sills are inferred to have been intruded at depths of 1–2 km into sedimentary rocks with TOC's ranging from 0.5–2%; as observed along the conjugate Møre and Vøring basins (Svensen et al., 2004; Aarnes et al.,

2015). Conversely, our observations from seismic data in the Upper Paleozoic Danmarkshavn Basin indicate the sills were emplaced at depths >3 km. This observation is supported by field and apatite-fission track data from the Upper Paleozoic-Mesozoic-aged basins of east Greenland, which indicate the sills are intruded at depths of >3 km into Jurassic-aged shales (Larsen and Marcussen, 1992). These shales have TOC contents ranging from 4–10% (Price and Whitham 1997), suggesting that the quantities of gas produced during contact metamorphism in the Danmarkshavn Basin were significantly higher than in Cretaceous and Paleocene basins elsewhere on the northeast Atlantic continental margin.

4.2 Quantities of gas produced

Following the method of Svensen et al. (2004), we calculate that the mass of methane (W_{CH4}) produced during contact metamorphism is: $W_{CH4} = 1.34F_C V_A \rho$, where 1.34 is the atomic weight conversion factor between carbon and methane, F_C is equal to the TOC content of the host rock, V_A is the volume of the aureole and ρ is rock density (2400 kg m⁻³). In our calculations we assume TOC contents of 4–10 wt. % in the Danmarkshavn Basin and 0.5–2 wt.% in the Thetis Basin. To calculate V_A we assume an aureole thickness of 100–600 m. This is based on our seismic data which indicates the sills are >>50 m thick, and the observation that aureole thickness for sills >50 m thick is equal to one sill thickness above and below the intrusion (Svensen et al., 2004). This sill to aureole thickness relationship is supported by many field and borehole studies globally (see review by Aarnes et al., 2010) and is supported by the closest analogue to the study area, where similar-aged intrusions of the Utgard sill complex were penetrated by well 6607/5-2 in the Vøring Basin (Aarnes et al., 2015). In the Utgard case, a ~1 km thick stratigraphic interval was demonstrated to be effected by the near simultaneous emplacement of two c. 100 m thick sills. Field data from east Greenland also supports our

interpretation, where sills typically range in thickness from 50–300 m (Larsen and Marcussen, 1992; Planke et al., 2005).

Based on these assumptions, our calculations indicate $0.06-1.73 \times 10^{18}$ g of methane was produced from the Danmarkshavn Basin and an additional $0.01-0.62 \times 10^{18}$ g of methane would have been produced as a result of intrusion emplacement in the Thetis Basin. The combined range for gas production along the northeast Greenland margin therefore equals $0.07-2.36 \times 10^{18}$ g of methane. These values represent 50–90% of the total gas production potential of the source rocks, since gas production depends on kerogen type (Hunt, 1996). Lower volumes of thermogenic gas may be produced during contact metamorphism as a result of heating of sediment pore water and trapping of volatiles (e.g. Gröcke et al., 2009). Gas volume production also varies as a result of host rock permeability, background temperature, aureole thermal profile and heat transfer mechanism (Aarnes et al., 2010; Iyer et al., 2013). However, we highlight that the main controls on the volume of gas produced are host rock composition, TOC, sill thickness and sill extent (Aarnes et al., 2011; Iyer et al., 2013) which we have accounted for in our estimates.

Additionally, there are several reasons for which we suggest that our estimate of gas production represents a conservative estimate. Firstly, many sills may be below the resolution of the seismic data (Schofield et al., 2015). Secondly, the calculated volumes do not include the poorly imaged regions beneath areas covered by extrusive volcanic rocks and areas with limited data coverage due to sea ice. Thirdly, the formation of hydrothermal vent complexes may have resulted in the breaching of pre-existing hydrocarbon reservoirs (e.g. Price & Whitham, 1997; Svensen et al., 2004) further adding to the potential quantities of gas released. Finally, our estimates do not include the contribution of magmatic CO₂ produced from degassing of the intrusions. It has been proposed that at the margin scale, volcanic CO₂ emissions are capable of complementing thermogenic gas release in perturbing the Earths'

atmosphere (Eldholm & Thomas, 1993; Saunders, 2016). Assessing the quantitative volumes of CO₂ released by intrusions is challenging without constraints on the volatile contents of the magmas emplaced into the Danmarkshavn and Thetis basins, however, such contributions could have increased the overall volume of released greenhouse gases supporting the conservative nature of our estimates.

4.3 Effect of gas release on the Paleocene carbon reservoir

To determine the potential climatic impact of thermogenic gas release, it is important to constrain the isotopic mass balance of the Paleocene carbon reservoir. To do this, we use the method described by Mc Inerney and Wing (2011). We assume that the initial mass of the Paleocene surface reservoir was 50×10^{18} g C, its carbon isotope ratio was -2.5% and that thermogenic methane has a δ^{13} C of $\sim -30\%$ (McInerney and Wing, 2011). We also use a value of -3.5% for the PETM initial isotope excursion in δ^{13} C (Zachos et al., 2007; Sluijs and Dickens, 2012). Based on these assumptions, we estimate that 2.5×10^{18} g CH₄ would have been required to cause the negative δ^{13} C excursion during the PETM. Our conservative estimates of gas production indicate that a comparable volume (2.36×10^{18} g) could have been produced along the northeast Greenland margin. A large proportion of these gases may have reached the atmosphere regardless of whether or not the eruptions occurred in the marine environment; numerical modelling shows that methane plumes formed during subaqueous gas eruptions do not become fully dissolved or oxidised in the ocean (Zhang, 2003).

5. Discussion

The absence of well data from the hydrothermal vents on the northeast Greenland margin prevents us from attaining the 10–100's ka resolution required to directly link vent formation to the PETM. Additionally, Iyer et al. (in review) document that not all generated gases will be

outgassed to the atmosphere within the short timescales associated with climate perturbations. However, the VH gives a robust relative time datum which is unequivocally associated with active venting across the northeast Greenland margin. This reflection can be clearly mapped laterally from the Thetis Basin to the SE where it terminates against the Inner Flows facies. A similar temporal relationship is observed for vent complexes on the Norwegian margin where the TV horizon, which represents the upper part of the vent complexes, terminates against the Inner Flows (Planke et al., 2005). Igneous intrusions, associated venting and the eruption of the Inner Flows on the Norwegian margin occurred during the onset of continental breakup at c. 55.6 Ma (Planke et al., 2005; Svensen et al., 2010). Given the conjugate nature of the two margins and the well-documented temporal and spatial association of the Inner Flows volcanism with continental break-up (Planke et al., 2000) it is highly likely that both the Inner Flows and venting occurred at similar times on either side of the developing rift system near the Paleocene-Eocene boundary. Our interpretation that venting occurred at this time is supported by radiometric dating of onshore volcanic rocks along the northeast Greenland margin which indicates that the main phase of volcanism occurred at 56-53 Ma (Larsen et al., 2014). Since hydrothermal vent complexes form within tens of years of sill intrusion (e.g. Jamtveit et al., 2004; Aarnes et al., 2010) the complexes identified within this study formed in close temporal proximity to the PETM which occurred at ~55.5 Ma (Westerhold et al., 2009). We also note that along the Norwegian margin, an additional $0.3-3.0 \times 10^{18}$ g of methane was produced as a result of sill emplacement (Svensen et al., 2004). Combined with the contribution from the northeast Greenland margin, this creates a combined volume of 0.37–5.5 $imes 10^{18}\,\mathrm{g}$ of methane produced by sill emplacement (Fig. 7). Additional sill complexes emplaced prior to the PETM are also briefly documented along the eastern Greenland margin (Larsen and Marcussen, 1992) and have been extensively mapped within the Faroe-Shetland Basin (Schofield et al., 2015). As detailed in this study, these sills were intruded into both Upper

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Paleozoic-Mesozoic and Cretaceous-Tertiary basins (Larsen and Marcussen, 1992; Schofield et al., 2015). Given the ubiquity of hydrothermal vent complexes now identified along the Norwegian and northeast Greenland margins, it is likely that the total volume of thermogenic methane produced during intrusion-induced metamorphism could have easily produced the negative δ^{13} C excursion observed during the PETM.

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6. Summary

Continental rifting is commonly associated with voluminous magmatism and perturbations in the Earth's climate. Our study uses newly acquired seismic data from the northeast Greenland margin to reveal the extent of two previously unconstrained offshore sill complexes emplaced during rifting of the Northeast Atlantic continental margin. Intrusion emplacement into shale-dominated sedimentary rocks with TOC contents of up to 10% resulted in rapid contact metamorphism and the production of up to 2.36×10^{18} g of methane. Much of this methane was released within tens of years of sill intrusion via a series of newly-discovered hydrothermal vent complexes. These vent complexes present the first evidence for thermogenic methane release along the northeast Greenland margin. The volume of gas produced along this margin alone is approaching that capable of causing the negative δ^{13} C excursion observed during the PETM. Combined with the volume of methane released from similar vents in the Vøring and Møre basins, and the ubiquity of similar-aged sill complexes elsewhere along the Norwegian and UK margins, we suggest that volcanically-induced greenhouse gas release was a common and important phenomenon along the northeast Atlantic margins in close proximity to the PETM. This study highlights and supports the important role of volcanic activity in driving global-scale climate change.

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Figures

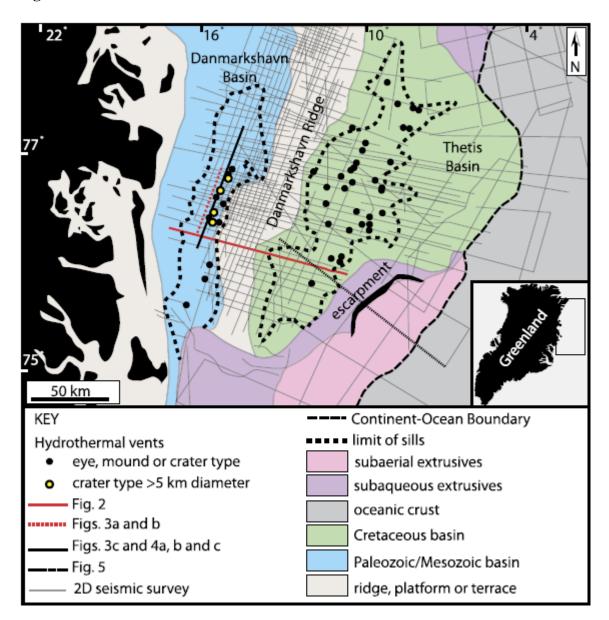


Figure 1. Map showing the Danmarkshavn and Thetis Basins and the distribution of volcanic units. Inset shows the location of the study area along the Greenland coast. Adapted from Hamann et al., 2005.

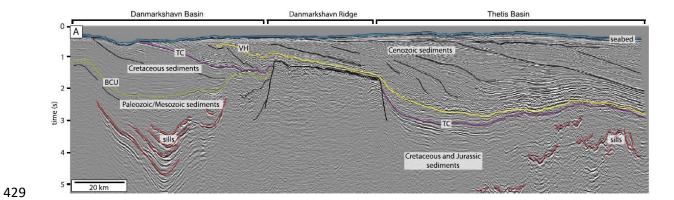


Figure 2. Seismic cross section showing sills within the Danmarkshavn and Thetis Basins. VH=Vent Horizon, TC=Top Cretaceous, BCU=Base Cretaceous Unconformity. Note that the sills in both basins are represented by white reflections, the same polarity as the seabed.

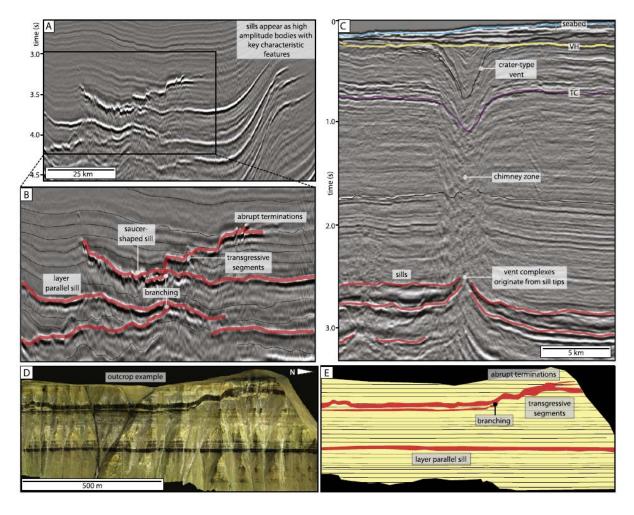


Figure 3. Diagram illustrating key features of sills. Images A and B are seismic cross sections of sills in the Danmarkshavn Basin. Note that the sills are high amplitude, positive reflections with layer parallel and saucer-shaped morphologies which transgress the stratigraphy. Image

C shows a crater-type vent linked to the tip of a sill within the Danmarkshavn Basin. See Figure 1 for location. Images D and E show field examples of sills from onshore Northeast Greenland with the same characteristics as the seismic examples.

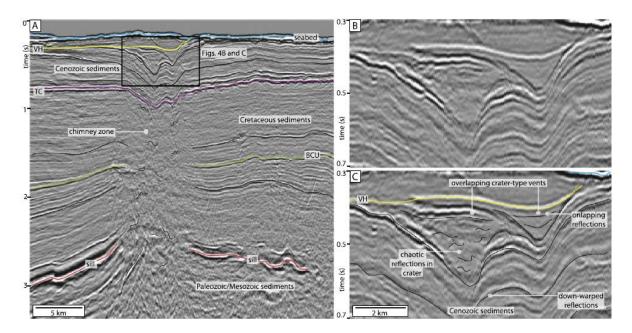


Figure 4. Seismic sections of crater-type hydrothermal vents above sills within the Danmarkshavn Basin. VH=Vent Horizon, TC=Top Cretaceous, BCU=Base Cretaceous Unconformity.

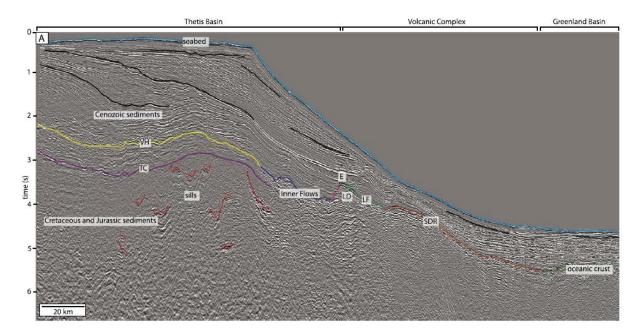


Figure 5. Seismic sections across the Thetis Basin showing the Vent Horizon (VH) terminating against the Inner Flows. SDR=Seaward Dipping Reflection, LF=Landward Flows, LD=Lava Delta, E=Escarpment.

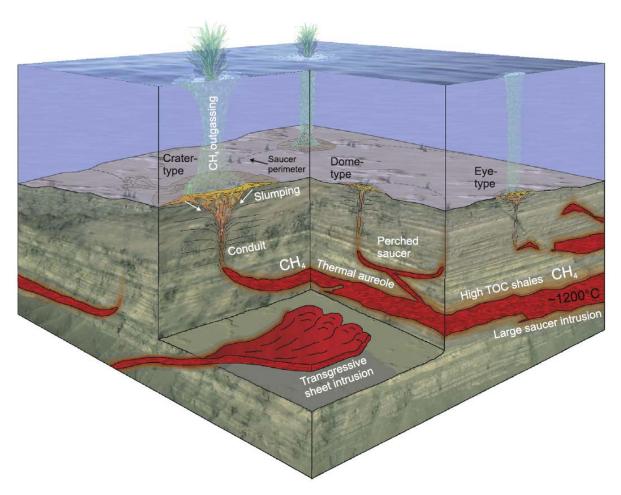


Figure 6. Schematic diagram illustrating the relationship of hydrothermal vents to underlying intrusions, and the subsequent release of CH₄.

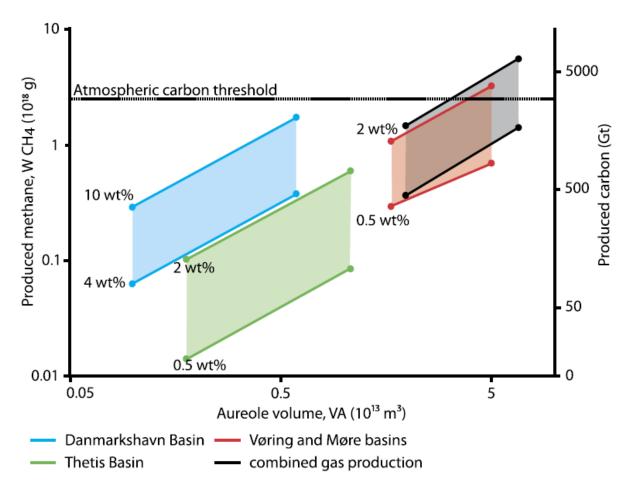


Figure 7. Graph showing estimated gas production from the Danmarkshavn, Thetis and Vøring and Møre basins. The black lines represent the minimum and maximum values for the combined gas production in these basins. The dashed line represents the threshold value of methane required to have produced the negative $\delta^{13}C$ excursion observed during the PETM. Gas production varies as a function of sill complex area, intrusion thickness and the TOC contents (shown in wt %) of the host sedimentary rocks.