

1 **Maturity Issues within Palaeocene Coal, Spitsbergen: Implications for local**  
2 **and regional burial and uplift models**

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13  
14 **The Central Tertiary Basin is an uplifted part of the North Barents Shelf and**  
15 **should be an ideal location to understand the thermal history, maximum**  
16 **burial depth and overburden thickness in this petroleum-rich area. Efforts to**  
17 **quantify the thermal history of the region have been hampered by reports of**  
18 **hyper-thermal conditions, maturity gaps and maturity inversions in the**  
19 **Tertiary vitrinite reflectance ( $R_o$ ) record. This has been attributed to thermal**  
20 **insulation effects, bitumen suppression and later Tertiary volcanism.**  
21 **Through the use of  $R_o$ , organic maturity parameters,  $^{13}C$  NMR and Rock-Eval**  
22 **pyrolysis, this study aims to explain the unusual maturity effects observed and**  
23 **the implications for burial models. Within single seams,  $R_o$  % ranges from 0.5-**  
24 **0.78 with increasingly bimodal distribution up-seam. Analysis of coal**  
25 **aromaticity and the results of Rock-Eval analysis confirm that maturity gaps**  
26 **and inversions only occur where the vitrinite reflectance has been suppressed**  
27 **by high bitumen content (300-400 mg/g coal). Samples with the lowest**  
28 **hydrogen index values (<250 mg HC/ TOC) provide the most accurate estimates**  
29 **of the vitrinite reflectance. Results indicate maximum burial temperatures of**  
30 **120°C in the basin centre and 100°C at the basin margins with a hyper-thermal**  
31 **gradient of approximately 50°C/km. This gradient implies a total overburden**  
32 **of 2 km of which 1 km has been lost. Maximum burial depth and total**  
33 **erosional sediment load to the Barents Shelf are therefore at the lower end of**  
34 **current estimates.**

35

36 **Key Words**

37 Vitrinite Reflectance, Oil prone coal, Maturity, Barent Shelf, Spitsbergen

38 **1. Introduction**

39 Apparent maturity gaps and maturity inversions in vitrinite reflectance ( $R_o$ ) data  
40 appear common within the Central Tertiary Basin, notably between the Triassic-  
41 Jurrassic boundary and Cretaceous-Basal Tertiary strata\_(Paech and Koch, 2001).  
42 Orheim et al. (2007) also report a maturity gap ( $R_o$  %=0.94 vs. 0.71) within Basal  
43 Tertiary Firkanten Formation coal seams which stratigraphically are only 40 m apart.  
44 Tertiary vitrinite reflectance ( $R_o$ ) data forms a key part of many estimates regarding  
45 geothermal gradient, maximum burial depth and overburden loss (Major and Nagy,  
46 1972, Manum and Throndsen, 1978, Throndsen, 1982, Paech and Koch, 2001). Any  
47 significant suppression of vitrinite may therefore lead to changes to estimates of erosion  
48 and transportation to local depocentres such as the Barents Sea,

49 Orheim et al., (2007) provide two possible explanations for apparent maturity variations  
50 within the Tertiary coals, namely the insulation effect of an underlying seam and  
51 suppression of vitrinite reflectance by bitumen enrichment. Hyper-thermal conditions  
52 during burial have been implied in a number of studies of the Adventdalen Area (Fig. 1;  
53 Major and Nagy, 1972, Manum and Throndsen, 1978, Throndsen, 1982, Paech and  
54 Koch, 2001, Braathen, 2012). In addition, it is clear that the Central Tertiary Basin was  
55 subject to local volcanic activity during the Tertiary as shown by numerous bentonite  
56 beds in Van Mijenfjorden Group sediments (Fig. 2), and a dolerite sill in the  
57 Bjørndalen/Fuglefjellet region\_(Pers. Comm. Trygvason Eliassen, 2014). The effect on  
58 vitrinite reflectance in the case of intrusions is expected to be significant but highly  
59 localised.

60 Vitrinite suppression by bitumen is well documented in oil source rocks including coals.  
61 Fluorescent (perhydrous) vitrinites enriched in hydrogen rich material are an indicator  
62 of oil potential and therefore of the potential for suppressed vitrinite reflectance ( $R_o$ )  
63 values (Diessel and Gammidge, 1998). As vitrinite reflectance is an indirect measure of  
64 aromaticity (Carr and Williamson, 1990) any excess aliphatic material will lead to  $R_o$   
65 values suppression. Orheim et al. (2007) observed that the upper Firkanten formation  
66 coals fluoresce under UV light and produce droplets of oil during preparation. In  
67 addition, Marshall (2013) show that the coals produce up to 40 wt% hydrocarbons, when

68 processed by Soxhlet solvent extraction and hydrous pyrolysis. This would indicate that  
69 the coals have at least some oil potential and therefore bitumen suppression may  
70 present a reasonable explanation.

71 In this study we examine bulk and high resolution vitrinite reflectance measurements to  
72 examine whether reported  $R_o$  variability can be replicated. We utilise other independent  
73 maturity parameters such as organic biomarkers, Rock-Eval and aromaticity to provide  
74 an alternative measure of maturity in the coals. Focussing upon the role of oil potential  
75 in  $R_o$  suppression we compare Rock-Eval hydrogen index (HI) values with vitrinite  
76 reflectance to attempt to correct for the suppression effect. The implications for the  
77 thermal regime, missing overburden estimates, sediment compaction and sediment  
78 transportation to the Barents Sea are then discussed.

## 79 **2. Geological Setting**

80 Svalbard (Fig.1) represents an uplifted part of the Northern Barents shelf (Harland et  
81 al., 1997) comprising Caledonian basement and subsequent unconformable basin infill.  
82 Much of central and southern Spitsbergen forms part of the Central Tertiary Basin a  
83 asymmetric synclinal basin, bounded to the east and west by the Billefjorden Faultzone  
84 and West Spitsbergen fold and thrust belt. Formed in response to the onset of  
85 compression related West Spitsbergen foreland fold and thrusting and prior to the  
86 strike-slip separation of Svalbard and Barents Shelf from Greenland (Harland, 1997,  
87 Tessensohn, 2001) the Central Tertiary Basin contains sediments dating from as early  
88 as the Carboniferous including many source rock analogues from the wider Barents sea  
89 region. It also contains the majority of the economic coal deposits on the islands, with  
90 mining concentrated within the Tertiary Firkanten Formation

91 In the NE Central Tertiary Basin, the Firkanten Formation comprises two sub-units;  
92 the lowermost Todalen Member and the overlying Endalen Member representing a  
93 sequence of paralic coalbearing tidal sediments overlain by laminated or heavily bio-  
94 turbated marine sandstones (Fig. 2; Dallmann *et al.*, 1999). The base of the Firkanten  
95 Formation is marked by a low angle unconformity with the Lower Cretaceous  
96 Carlinefjellet formation, sometimes marked by a basal conglomerate known as the  
97 Grønfjorden bed (Harland, 1997, Dallmann, 1999).

98

99 To the west an additional offshore unit is also observed, known as the Kolthoffberget  
100 Member (Fig. 2; Dallmann, 1999). The Todalen Member represents the main coal

101 bearing unit within the Van Mijenfjorden Group and therefore is the focus of this study.  
102 It consists of 3-5 siltstone-sandstone-coal successions representing increased subsidence  
103 and the infilling of the Cretaceous pene-plain (Harland, 1997, Dallmann, 1999). Five  
104 main coal seams are commonly cited within the Todalen Member; the Svea, Todalen,  
105 Longyear, Svarteper and Askeladden Seams (Fig. 2; Dallmann, 1999, Harland, 1997,  
106 Orheim et al., 2007)).

107

### 108 3. Methodology

109 Coal was sampled (Fig. 1) from the Svea Nord, Longyear and Svarteper seams from  
110 mine sections in Svea Nord and Mine 7, boreholes from the Lunckefjellet (BH6A-2007,  
111 BH10/2007, BH10/2009, BH15/2011), Adventdalen (BH4/2009, BH5/2009) and  
112 Colesdalen (BH3/2008) regions, and a field section from Bassen. Samples were coarse-  
113 crushed, separated by cone and quarter (to allow unbiased sampling for coal maceral  
114 analysis) and the remainder fine crushed (<100  $\mu\text{m}$ ). Polished blocks (particle size; 0.2 –  
115 1 mm) were created for organic petrology and vitrinite reflectance ( $R_o$ ).

116

117 Coal vitrinite reflectance ( $R_o$ ) was determined using a microscope fitted with a 50x oil  
118 immersion objective and 10x oculars, 12 V 100 W quartz halogen lamp and 100 W HBO  
119 high pressure mercury lamp.  $R_o$  measurements were taken using an attached  
120 photomultiplier (100 points) and calibrated using a 1.24  $R_o$ , 564 nm glass prisma  
121 standard and blank plastic oil-filled depression according to BSI standards (British  
122 Standard Institution, 2009).

123

124 To determine the amount of hydrocarbon potential the coals were analysed using a  
125 Rock-Eval6 analyser configured in standard mode (pyrolysis and oxidation as a serial  
126 process) to indicate the coals (20 mg dry wt) were heated from 300°C to 650°C at  
127 25°C/min in an inert atmosphere of  $\text{N}_2$  and the residual carbon then oxidised at 300°C to  
128 850°C at 20°C/min (hold 5 min). Hydrocarbons released during the two stage pyrolysis  
129 were measured using a flame ionization detector and CO and CO<sub>2</sub> measured using an IR  
130 cell. The performance of the instrument was checked every 10 samples against the  
131 accepted values of the Institut Français du Pétrole (IFP) standard (IFP 160 000, S/N1 5-  
132 081840). Classical Rock-Eval parameters were calculated by integration of the amounts  
133 of HC (thermo-vaporized free hydrocarbons) expressed in mg/HC/g rock ( $S_1$ ) and  
134 hydrocarbons released from cracking of bound OM expressed in mg/HC/g rock ( $S_2$ )

135 (Engelhart et al., 2013). The Hydrogen Index (HI) was calculated from  $S_2 \times 100/\text{TOC}$  and  
136 the Oxygen Index (OI),  $S_3 \times 100/\text{TOC}$ . The error on the  $T_{\text{max}}$  is about  $\pm 6$  °C.

137

138 In order to examine organic maturity parameters in the solvent extractable hydrocarbon  
139 fraction from the Adventdalen coals (BH-5-2009 and Mine 7 Section), the coals were  
140 subjected to accelerated solvent extraction (ASE) using a 93:7 DCM: methanol mixture  
141 for a period of 24hrs, and separated into aliphatic, aromatic and polar fractions by silica-  
142 alumina adsorption column chromatography (15 ml *n*-hexane; 15 ml *n*-hexane:DCM (3:2  
143 v/v); 15 ml DCM:,methanol (1:1 v/v). Aliphatic and aromatic fractions were analysed by  
144 GC-MS in both the SIM and full scan ( $m/z$  50-450) modes, using a Varian CP-3800 gas  
145 chromatograph, interfaced to a Varian 1200 mass spectrometer (EI mode, 70eV).  
146 Separation was achieved on a VF-1MS fused silica capillary column (50m x 0.32 mm i.d,  
147 0.25  $\mu\text{m}$  stationary phase thickness), with helium as the carrier gas, and an oven  
148 programme of 50°C (hold for 2 min) to 300°C (hold for 20.5 min) at a heating rate of  
149 4°C/min. The  $m/z$  85 single ion chromatogram (SIC) was used to measure *n*-alkane peak  
150 area response. Relative hopane and sterane concentrations were from peak area  
151 responses in the  $m/z$  191 and  $m/z$  217 SICs, respectively. MPI-1 was calculated from  
152 the aromatic fraction from peak area responses from the  $m/z$  178 and  $m/z$  192 SICs  
153 | respectively (after Cassani et al., 1988).

154

155 The ratio of aliphatic to aromatic components was determined in selected Svea Nord and  
156 Longyear coal samples (1.20 m and 1.30 m above seam base respectively) using high  
157 resolution solid state 50 MHz  $^{13}\text{C}$  NMR. Analysis was carried out in a BrukerAvance  
158 200 spectrometer to ascertain using the cross polarisation (CP) sequence in conjunction  
159 with magic angle spinning (MAS). For CP-MAS analysis, the acquisition time was 0.05  
160 s, the relaxation delay was 1.5 s and the contact time was 1 ms. Samples were packed  
161 tight into a cylindrical (7 mm o.d.) zirconia rotor with a cap made of a homopolymer of  
162 chlorotrifluoroethene (Kel-F) and spun at the magic angle (54.74°) with a spinning rate  
163 of approximately 5 kHz. Tetrakis-trimethylsilyl silane (TKS) was added to the samples  
164 as an internal standard. The number of scans was 2500 and the free induction decays  
165 (FIDs) were processed using a line broadening factor of 50 Hz.

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## 170 4. Results

### 171 4.1 Vitrinite Reflectance

172 Examination of vitrinite reflectance values (Table 1) from two bulk samples of Svea and  
173 Longyear coal examined replicate the maturity gap reported by Orheim et al. (2007)  
174 with  $R_o$  % values of 0.65 vs 0.78. Consequently, to understand how widespread and the  
175 main cause of this difference in maturity the Longyear seam was examined at higher  
176 resolution in coals from three sub-regions; Adventdalen, Lunckefjellet and Colesdalen.

### 177 4.2 Adventdalen

178 Coals from the Adventdalen region are vitrinite dominated with low inertinite and  
179 liptinite with the exception of the Svea seam which is inertinite dominated. Vitrinites  
180 are observed to exhibit dull orange/brown fluorescence under blue light.  $R_o$  values range  
181 from 0.5-0.8 with the Svea seam showing generally highest values and uppermost  
182 Longyear seam the lowest (Fig 3; Table S1). All sites show a similar range of  $R_o$  values  
183 which is unexpected as the Bassen section is up-dip of samples from Breinosa area. The  
184 distribution of  $R_o$  values exhibits the greatest range in Mine 7 and Bassen samples with  
185 minimum values at Mine 7 of 0.5. There are distinct differences between upper and  
186 lower seams in Mine 7 and Bassen section. These numerous small and large scale  
187 variations comprising rapid  $R_o$  drops and inversions are unlikely to be the product of  
188 differing thermal histories. Conversely  $R_o$  values from the BH4/2009 sample locality  
189 remain consistently low, which is perhaps the product of a more homogenous  
190 composition.

191 When the distribution of  $R_o$  data in the Mine 7 section is examined in greater detail it  
192 indicates a general broadening of  $R_o$  measurements up-seam (Fig. 4) accompanied by a  
193 gradual reduction in the number of higher reflectance measurements. In addition  
194 rather than a single main peak, measurements become increasingly bimodal up-seam.  
195 This bi-modality is possibly the product of variation in the relative number of  
196 fluorescent vs. non-fluorescent vitrinites.

### 197 4.3 Lunckefjellet

198 The Lunckefjellet Longyear seam is vitrinite dominated (as in Adventdalen) with low  
199 inertinite and liptinite. Vitrinites again exhibit dull orange/brown fluoresce under blue  
200 light. Highest vitrinite reflectance values ( $\approx 0.76$ ) are found towards the eastern margin

201 of the basin (Fig 5; Table S1) with decreasing  $R_o$  down dip which is contrary to  
202 expectations. In addition the Longyear seams exhibit a general decrease in  $R_o$  up-seam  
203 similar to that seen in Adventdalen. The greatest range of  $R_o$  values (Fig. 5) can be found  
204 in the easternmost sampling locality's with values ranging from  $R_o$  % 0.59-0.76. These  
205 numerous small and large scale variations comprising rapid  $R_o$  drops and inversions are  
206 unlikely to be the product of differing thermal histories. As in Adventdalen, when the  
207 distribution of  $R_o$  data in the Lunckefjellet section (Fig. 6) is examined in greater detail  
208 it indicates a general broadening of  $R_o$  measurements up-seam (Fig 6). In addition  
209 measurements become increasingly bimodal up-seam. The high degree of similarity  
210 between the Adventdalen and Lunckefjellet coals indicates that a similar process is  
211 controlling the large variations in vitrinite reflectance at both sites.

#### 212 4.4 Colesdalen

213 The Colesdalen coals are higher in inertinite (compared to the other coals in this study)  
214 but still vitrinite dominated. The Colesdalen coals have elevated  $R_o$  values (0.78) as  
215 expected from a more central part of basin (Table S1). In addition the Colesdalen coals  
216 show less variability with values consistently high throughout the seam with the  
217 exception of two more ash rich samples.

218 The vitrinite reflectance measurements (Table S1) from the Lunckefjellet and  
219 Adventdalen regions show large variations ( $R_o$  %  $\approx$  0.3) within some parts of the seam  
220 and importantly replicate the maturity gap observed (Orheim et al., 2007). These  
221 numerous small and large scale variations comprising rapid  $R_o$  drops and inversions are  
222 unlikely to be the product of differing thermal histories. As a result, the suitability of  
223 vitrinite reflectance in these coals as a measure of maturity will be examined by  
224 comparing  $R_o$  data with independent measures of maturity such as organic maturity  
225 parameters, Rock-Eval and aromaticity.

#### 226 4.5 Organic Maturity parameters

227 Coals from BH 5/2009 and Mine 7 were selected to further examine the true maturity of  
228 the Svea, Longyear and Svarteper seams through organic geochemical biomarkers in  
229 Soxhlet extracted oils. Organic Geochemical maturity parameters (Table 1) can be  
230 highly specific at low maturities but often reach equilibrium at around  $R_o$  0.7%, making  
231 many of little use at higher maturities (Peters et al., 2005).

232 Aliphatic maturity parameters for the Todalen coals are at or approaching equilibrium  
233 (Table. 1) indicating a maturity in excess of  $R_o$  0.7%. The *n*-alkanes show little odd over  
234 even predominance, with carbon preference index values (CPI(1),(Bray and Evans,  
235 1961)) close to mature ratios ( $\approx 1$ ). Notably, both CPI(1), sterane and hopane maturity  
236 parameters shown no change up-seam in the Longyear (Fig.7) contrary to what would be  
237 expected if  $R_o$  values at the top of the seam represented true maturity values and were  
238 not suppressed.

239 Consideration of the aromatic Methylphenanthrene Index, MPI-1 (Radke et al., 1982,  
240 Radke et al., 1986, Radke, 1988) predicts  $R_o$  values of around  $0.72 \pm 0.05$  in the Svea,  
241 Longyear and Svarteper seams in the Adventdalen region with no changes up-seam  
242 (Table 1). Although the predicted values are lower than maximum  $R_o$  values measured  
243 in these coals ( $R_o \% \approx 0.78$ ), the predicted values remain substantially higher than the  
244 lowest values measured at the top of the Longyear seam in Mine 7 ( $R_o \% \approx 0.5$ ). A  
245 possible explanation for the difference is the effect of variation in organic matter source  
246 and migration (Peters et al., 2005). Differences between predicted MPI-1  $R_o$  % (0.72)  
247 and measured  $R_o$  (0.79) in the Svea seam at both Adventdalen and Svea Nord are likely  
248 a product of differing palaeo-environment and associated organic matter.

249 In summary, geochemical evidence points to both seams having comparable maturities  
250 and thermal histories. Significantly, the  $R_o$  maturity gap, both within the Longyear  
251 seam and between the Svea and Longyear seams is not reproduced geochemically.

#### 252 **4.6 Oil Potential and Maturity**

253  $T_{max}$ , like many maturity parameters is highly dependent upon source material (Peters  
254 et al., 2005) but is considered of use for assessment of Type II material (420-460°C) and  
255 Type III (400-600°C; Tissot et al., 1987).  $T_{max}$  ranges from 425–448°C for all the  
256 Svalbard coals indicating a maximum maturity in the early-mid oil window (Table. S2).  
257 When  $T_{max}$  is plotted against HI (indicative of source rock potential; Fig. 8) there is a  
258 general positive correlation with coal from the basin margin at Lunckefjellet and Bassen  
259 least mature and Colesdalen coals from the basin centre more mature. HI also appears  
260 to peak within the coals between  $T_{max}$  values between 435 and 445°C. This is similar to  
261 the  $HI_{max}$  concept, and is associated with the reorganisation of kerogen structure  
262 (Petersen, 2005).



263 | When  $T_{\max}$  is converted to  $R_o$  (Teichmüller and Durand, 1983) values were elevated  
264 compared to measured values indicating thermal maturity  $R_o$  %  $\approx 0.83$  (Table S2) for all  
265 samples in the Adventdalen region with the exception of Bassen where the more  
266 marginal basin setting means the coals are inherently less mature. Coals from the Svea  
267 Nord seam are slightly more mature than those in Adventdalen, consistent with the  
268 southward tilting of the Central Tertiary Basin. The Svea Nord seam has values around  
269 0.85. The Lunckefjellet coals appear less mature than the Svea Nord and Adventdalen  
270 coals with calculated  $R_o$  % values of 0.66, perhaps consistent with a more marginal  
271 setting. The Colesdalen coals as expected due to their more central location are more  
272 mature with calculated  $R_o$  % values of 0.83.

273 The production index ( $PI = S_1/(S_1+S_2)$ ) is another indicator of maturity values,  $< 0.1$   
274 | indicate an immature source rock and values  $> 0.4$ , a mature source rock (Maky and  
275 Ramadan, 2008). The Svalbard coals all have values  $< 0.1$  (Table. S2; Fig 8), suggesting  
276 the coals were exhumed prior to any significant generation of hydrocarbons, with  
277 Colesdalen the most mature with values approaching 0.1 (Fig.8). As with  $T_{\max}$  the coals  
278 with the highest HI values appear to be closest to generation, particularly in the  
279 Colesdalen region (Fig.8). The Bassen region appears to have unusually low free  
280 hydrocarbon values which may reflect weathering at the field site. Values of  $T_{\max}$   
281 appear slightly high compared to PI values which indicate that the coals are sub-  
282 mature. This may be due to the extended oil window in coals compared to other  
283 conventional source rocks (Petersen and Nytoft, 2006).

284 HI values in the Svea seam are generally  $< 250$  mg HC/TOC with upper seams ranging  
285 between 250-400 mg HC/TOC (Table S2). This confirms that the upper coal seams  
286 across eastern central basin have significant oil potential compared to the Svea Seam.  
287 The plot of HI vs OI (Fig.8) shows that most samples have compositions between that of  
288 Type II and Type III kerogen. OI is elevated and HI lower in the Bassen area indicates  
289 either lower maturity or weathering of samples. As this is a field sample and the  
290 vitrinite reflectance variations are similar to other sample locations in the area it is  
291 thought most likely to indicate weathering.

292 Examination of the relationship between HI and  $R_o$  (Fig.8) shows that all sites show  
293 strong negative correlation between  $R_o$  and HI indicating that the higher the oil  
294 potential the more suppressed vitrinite becomes. However the different gradients and  
295 positions of these lines show that this relationship is complicated by other factors.  
296 These are likely to be compositional, positional and weathering effects. This indicates

297 that samples with the lowest HI values provide a better reflection of the degree of  
298 coalification ( $\approx R_o$  % 0.78-0.80). This fits better with the production and migration of  
299 hydrocarbons described previously (Orheim et al., 2007, Marshall, 2013). Consequently,  
300 bitumen must have a suppressing effect on  $R_o$  across the basin.

#### 301 **4.7 Quantifying the suppression effect**

302 Direct measurement of aromaticity (of which  $R_o$  is an indirect measurement) is a useful  
303 tool in the derivation of maturity in some coals (Stephens et al., 1985, Carr and  
304 Williamson, 1990).

305 The aromaticity (%) of the Svalbard coals (Fig.9) differs greatly between the Longyear  
306 and Svea seams (50% vs. 70% respectively). Using the calibration of Carr and  
307 Williamson, (1990), yields equivalent  $R_o$  values of 0.76 % (Svea) and 0.50% (Longyear).  
308 This is clearly not the case, as the Longyear seam bears no resemblance to a brown coal  
309 and is likely to have entered the early oil window in most areas. The Svea, which is not  
310 oil prone, exhibits an aromaticity consistent with observed maturity. Consequently, the  
311 observed maturity gap between the Svea and Longyear seams must be caused by  
312 significant amounts of additional aliphatic material within the Longyear seam.

313 To quantify this suppression effect from normal bituminous Svea coal (75 parts  
314 aromatic: 25 parts aliphatic), additional aliphatic carbon within the oil prone Longyear  
315 coal would account for 33% of total carbon. This is approximately equivalent to 380-  
316 400mg/g TOC, which is very close to HI values observed throughout the Eastern Central  
317 Tertiary Basin (Table S2). Variations in the amount of additional aliphatic material  
318 must therefore be responsible for apparent inter and intra seam maturity variation,  
319 maturity gaps and maturity inversions within Firkanten Formation coals.

## 320 **5. Discussion**

### 321 **5.1 Maturity of coals**

322 At a bulk scale the  $R_o$  difference observed (Orheim et al., 2007) has been replicated with  
323 the Longyear seam in particular appearing to show a rapid decrease in maturity up-  
324 seam. However, this variability does not appear to be replicated by independent  
325 measures of maturity such as organic maturity parameters and Rock-Eval. In addition  
326 these parameters indicate that the coals have a thermal maturity in excess of  $R_o$  % 0.70  
327 indicating that the coals reached maturities consistent with the early to mid oil window.

328 The bulk sample from the Longyear coal contains around 30% extra aliphatic material  
 329 compared to the Svea seam. This is likely to reflect enrichment in bitumen. This liquid  
 330 hydrocarbon potential is seen throughout the localities with Rock-Eval HI values  
 331 ranging from 250-400mg HC/TOC. This is consistent with observed oil production  
 332 (Orheim et al., 2007) and total hydrocarbon yields from Rock-Eval (300-400 mg  
 333 HC/TOC) and Soxhlet/hydrous pyrolysis (300-400 mg/g HC; Marshall, 2013).

334 The effect upon  $R_o$  is clear (Fig.8) showing the higher HI values are the more suppressed  
 335  $R_o$  values become. This vitrinite suppression effect by the enrichment of later seams in  
 336 aliphatic rich bitumen compared to non-oil prone Svea seams is therefore considered the  
 337 primary cause for the maturity gaps and inversions observed by Orheim et al., (1997)  
 338 (Orheim et al., 2007). Consequently, earlier Carboniferous, Jurassic and Cretaceous  
 339 coals in the basin with observed coal rank inversion and maturity gaps (Paech and  
 340 Koch, 2001) may benefit from further examination of this effect. The most accurate  
 341 values for the Longyear seam appear to be found at the base of the seam which would  
 342 give thermal maturities of the coals of  $R_o$  % of 0.78 at Breinosa, 0.68 at Bassen, 0.76 at  
 343 Lunckefjellet and 0.80 in the Colesdalen area.

## 344 5.2 Thermal regime and implications for overburden models

345 Previous models measuring coalification gradients in Tertiary strata from the  
 346 Adventdalen area range from between 0.17-0.32%  $R_o$ /km (Paech and Koch, 2001 and  
 347 references therein). However due to the bitumen suppression effect it is likely that  $R_o$   
 348 values lie somewhat higher than previously thought. As the highest values of  $R_o$  in each  
 349 seam appear the most reliable the maximum overburden and thermal regime at peak  
 350 burial was calculated using these values and the empirical palaeo-temperature equation  
 351 (Barker and Pawlewicz, 1994);

$$\text{Max T (}^\circ\text{C)} = \frac{\text{Ln}(R_o) + 1.68}{0.0124}$$

352  $R_o$  values indicate estimated peak temps of 116°C in the Colesdalen area, 111°C at  
 353 Breinosa, 110°C at Lunckefjellet and 100°C at Bassen. As expected, coals from the  
 354 centre of the basin were exposed to the highest temperatures and vice versa. Given  
 355 normal continental geothermal gradients (25°C/km) and assuming no other heat sources  
 356 (Corcoran and Clayton, 1999) this would reflect maximum burial depths of 4.6 km, 4.4  
 357 km, 4.4 km and 4.0 km respectively. In the Adventdalen area this would equate to a

358 total missing overburden of 3.4 km which is much greater than other estimates from the  
359 Central basin (Paech and Koch, 2001 and references therein).

360 Assuming reported  $R_o$  values of 0.43 for the upper Tertiary coals from the Aspelintoppen  
361 Formation are correct (Thronnsen, 1982) and values for the stratigraphically lower  
362 Firkanten Formation coals 0.78 then the  $R_o$  gradient will be 0.35  $R_o/km$ . This is  
363 consistent with the highest previous estimates (Paech and Koch, 2001). When converted  
364 using the Barker and Pawlewicz, 1994 equation this equates to a geothermal gradient of  
365 around 50°C/km, similar to previous estimates (40°C/km; Braathen et al., 2012) and  
366 maximum burial depths of 2.3km in the Colesdalen area, 2.2km at Breinosa, 2.2 km at  
367 Lunckefjellet and 2.0 km at Bassen. As approximately 1.0 km of overlying strata still  
368 exists in the Adventdalen Region (Thronnsen, 1982) the missing overburden is likely to  
369 amount to ~1.0 km. This is lower than the values of 1.7km previously reported (Paech  
370 and Koch, 2001 and references therein). In more central and southern areas of the  
371 basin, accounting for vitrinite suppression, would also greatly reduce the current  
372 estimated 3 km of eroded overburden (Manum and Thronnsen, 1978).

373 Previous studies support the conclusion (Major and Nagy, 1972, Manum and Thronnsen,  
374 1978, Thronnsen, 1982, Paech and Koch, 2001) that the geothermal gradient during  
375 burial was hyper-thermal. This is unsurprising as the Central Tertiary Basin was  
376 formed during a period of both local and regional tectonic activity. Additionally the  
377 presence of a dolerite sill in the Bjørndalen area (Pers. Comm. Trygvason Eliassen,  
378 2014) and the numerous volcanic bentonites within Tertiary strata indicate volcanism  
379 played an important role in shaping the thermal history of the Central Tertiary Basin.  
380 Hyper-thermal conditions may also explain the relatively low levels of compaction  
381 observed in Tertiary strata when compared to their apparent maturity.

382 Within a regional context, the Northern Barents including Svalbard (regarded as an  
383 uplifted NW edge of the Barents Shelf, Harland, 1997) was subjected to less burial and  
384 subsequent uplift and erosion than previously thought (Manum and Thronnsen, 1978,  
385 Thronnsen, 1982, Paech and Koch, 2001) and consequently fits with lower estimates of  
386 erosion and sediment load to the Southern Barents during the late Cenozoic (Rasmussen  
387 and Fjeldskaar, 1996).

388

389

390 **6. Conclusions**

391 Inferred maturity gaps within the Svalbard coal are shown to be due to suppression of  
 392 vitrinite reflectance by bitumen enrichment. Upper Firkanten formation coals have  
 393 significant oil potential (HI 250-400mg HC/TOC), they appear most affected by maturity  
 394 issues, displaying considerable variability in  $R_o$  values. Only the lowermost (and least oil  
 395 prone) parts of the coal seams remain relatively unaffected. Using these values for the  
 396 Adventdalen region, indicates  $R_o$  % values ranging from 0.68 at the basin margin to 0.78  
 397 4 km down-dip, At Lunckefjellet  $R_o$  % values are around 0.76 and in the more centrally  
 398 located Colesdalen values of 0.80. Coalification gradients in Adventdalen equate to  
 399  $\sim 0.35 R_o/km$  consistent with highest previous estimates (Paech and Koch, 2001), a  
 400 thermal gradient of approximately  $50^\circ C/km$  and peak burial depths of  $\sim 2km$  in the  
 401 Adventdalen region. This indicates overall overburden erosion was less than previous  
 402 estimates (1.7km; Manum and Throndsen, 1978) at around 1.0 km. The results of this  
 403 study indicate that burial and subsequent uplift and erosion were perhaps lower than  
 404 previously thought, leading to less compaction of tertiary strata on Svalbard and  
 405 resulting in reduced sediment load to the Barents Shelf in the Late Tertiary.

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528 **Figure and Table Captions**

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530 **All figures meant for colour online only**

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532 **Table 1** – Vitrinite Reflectance ( $R_o$ ), *n*-alkane, hopane, sterane and aromatic biomarker  
 533 and maturity parameters within the Central Tertiary Basin coals. <sup>a</sup> after Cassani  
 534 *et al.*, (1988), <sup>b</sup> after Radke *et al.*, (1982) *Pr/Ph* –*pristane/phytane*,  $Pr/nC17 =$   
 535 *Pristane/heptadecane* ratio,  $CPI(1) = \frac{2(C_{23}+C_{25}+C_{27}+C_{29})}{(C_{22}+2(C_{24}+C_{26}+C_{28})+C_{30})}$ ,  $TS/TM = 17\alpha-22,29,30\text{-trisorhopane} / 18\alpha-22,29,30\text{-}$   
 536 *trisorhopane*,  $TS/TH = \text{Total hopanes} / \text{Total steranes}$   
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539

540 **Table S1** –Vitrinite Reflectance ( $R_o$ ) values and location for all sites used in this study

541

542 **Table S2** – Rock-Eval Pyrolysis results for all sites used in this study.  $T_{max}$  conversion  
 543 to predicted  $R_o$  after *Teichmüller and Durand, 1983* and max burial depth  
 544 calculated from vitrinite reflectance data after *Barker and Pawlewicz, 1994*  
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548 **Figure 1** – Map of the island of Spitsbergen showing the mines and settlements of the  
 549 NW Central Tertiary Basin and sample locality and type.

550

551 **Figure 2** – The Van Mijenfjorden Group representing sedimentary infill of the Central  
 552 Tertiary Basin, modified from Helland-Hansen (1992)

553

554 **Figure 3** – Comparison of the vitrinite reflectance profile in the Longyear Seam across  
 555 Adventdalen from (A) BH4/2009 (B) Mine 7 Section (C) Bassen Field Section.  
 556 Note the large variations seen in rank and the general trend to lower  $R_o$  % values  
 557 upseam

558

559 **Figure 4** – Variation in rank ( $R_o$ ) within the Longyear seam in Mine 7, Adventdalen.  
 560 Examination of the distribution of vitrinite reflectance values measured shows a  
 561 shift to lower values, bi-modal distribution upseam.

562

563 **Figure 5** – Comparison of the vitrinite reflectance profile in the Longyear Seam across  
 564 Lunckefjellet from (A) BH 6A/2007 (B) BH 10/2007 (C) BH 15/2011 (D) BH  
 565 10/2009. Note the large variations seen in rank and the general trend to lower  
 566  $R_o$  % values upseam

567

568 **Figure 6** – Variation in rank ( $R_o$ ) within the Longyear seam in BH15-2011,  
 569 Lunckefjellet. As in Adventdalen examination of the distribution of vitrinite  
 570 reflectance values measured shows a shift to lower values, bi-modal distribution  
 571 upseam.

572

573 **Figure 7** – Variation in hopane and *n*-alkane maturity parameters up-seam within the  
 574 Longyear seam. Note little variation in maturity up-seam indicating no maturity gap.

575

576 **Figure 8** – Rock-Eval data from the Svalbard coals shows (A) HI vs  $R_o$  % shows that HI  
 577 (oil potential exerts a strong control upon vitrinite reflectance in all areas studied  
 578 N.B ash rich samples were removed as not representative and Lunckefjellet  
 579 sample represents BH15/2011. (B) HI vs OI shows that the composition of the



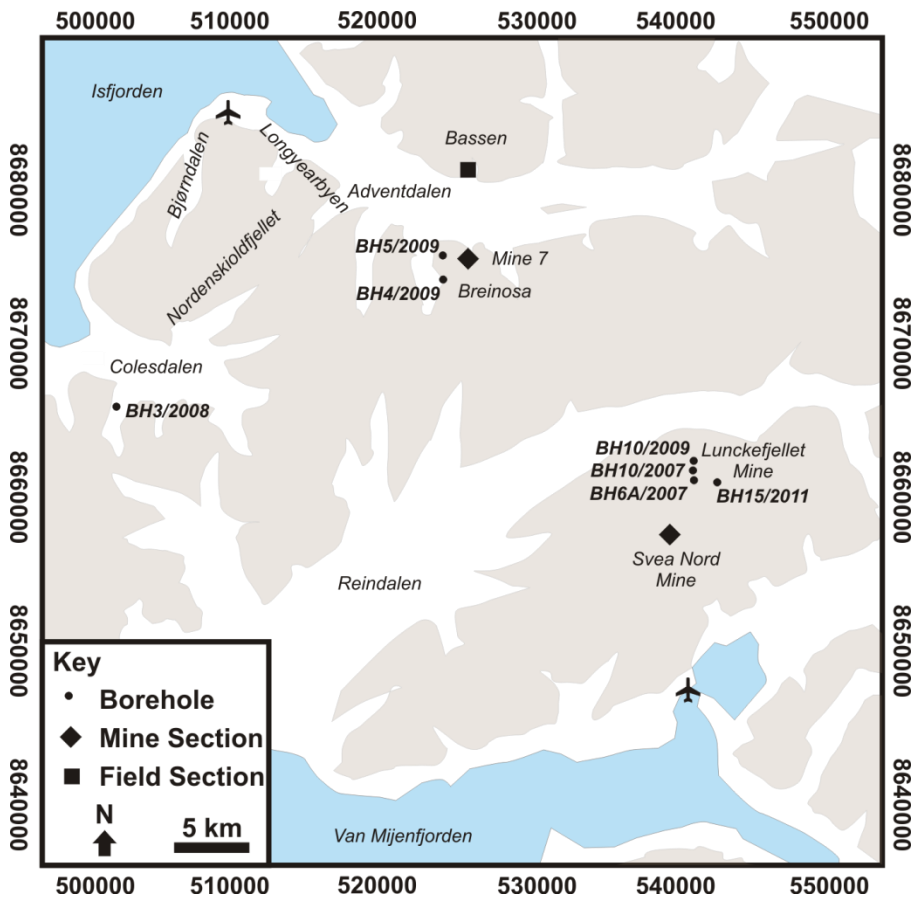
580 Svalbard coals is a cross between Type II and Type III kerogen with the  
581 exception of Bassen Samples which has elevated OI values likely due to  
582 weathering effects. (C) HI vs  $T_{max}$  and (D) HI vs. PI show that the Colesdalen  
583 coals are most mature and closest to hydrocarbon generation with the least  
584 mature coals found at Bassen. This is expected as Colesdalen is located in a  
585 more central basinal position than Bassen at the basin margins.

586 **Figure 9** – Measurement of aromaticity of the Svalbard Coals of by  $^{13}C$  NMR (A)  
587 Longyear seam and (B) Svea seam. Note the large difference between the seam  
588 indicating an apparent maturity equivalent to that of R0 0.5% and 0.78%  
589 respectively (Carr and Williamson, 1990). This cannot be the case as the  
590 Longyear is not a lignite, therefore the Longyear is enriched with non-coaly  
591 aliphatics.

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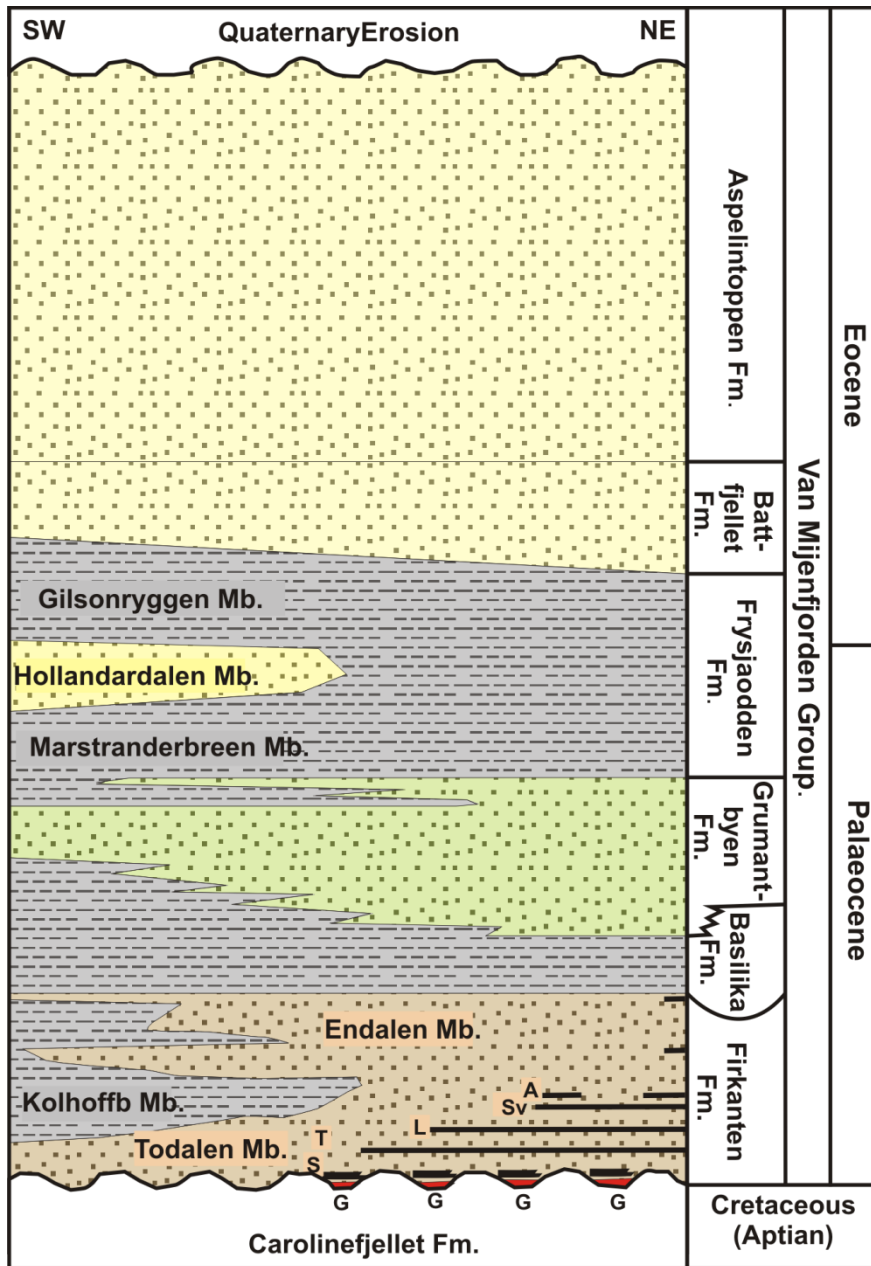
Figure 1



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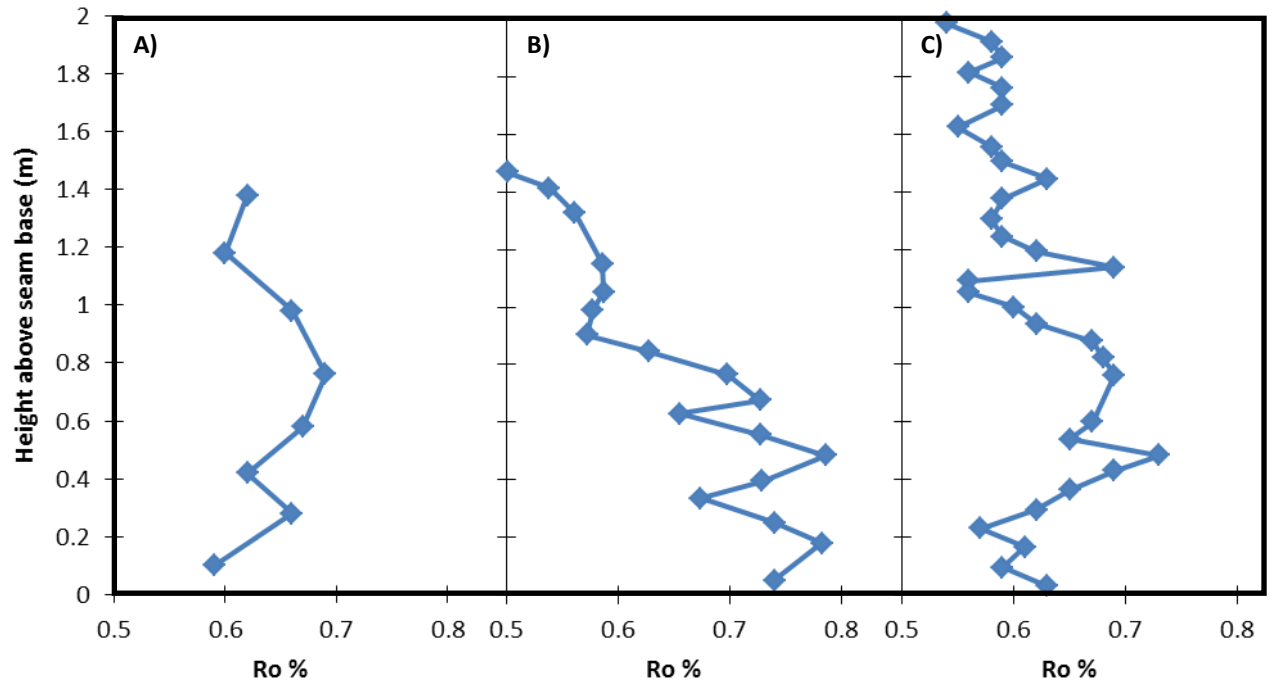
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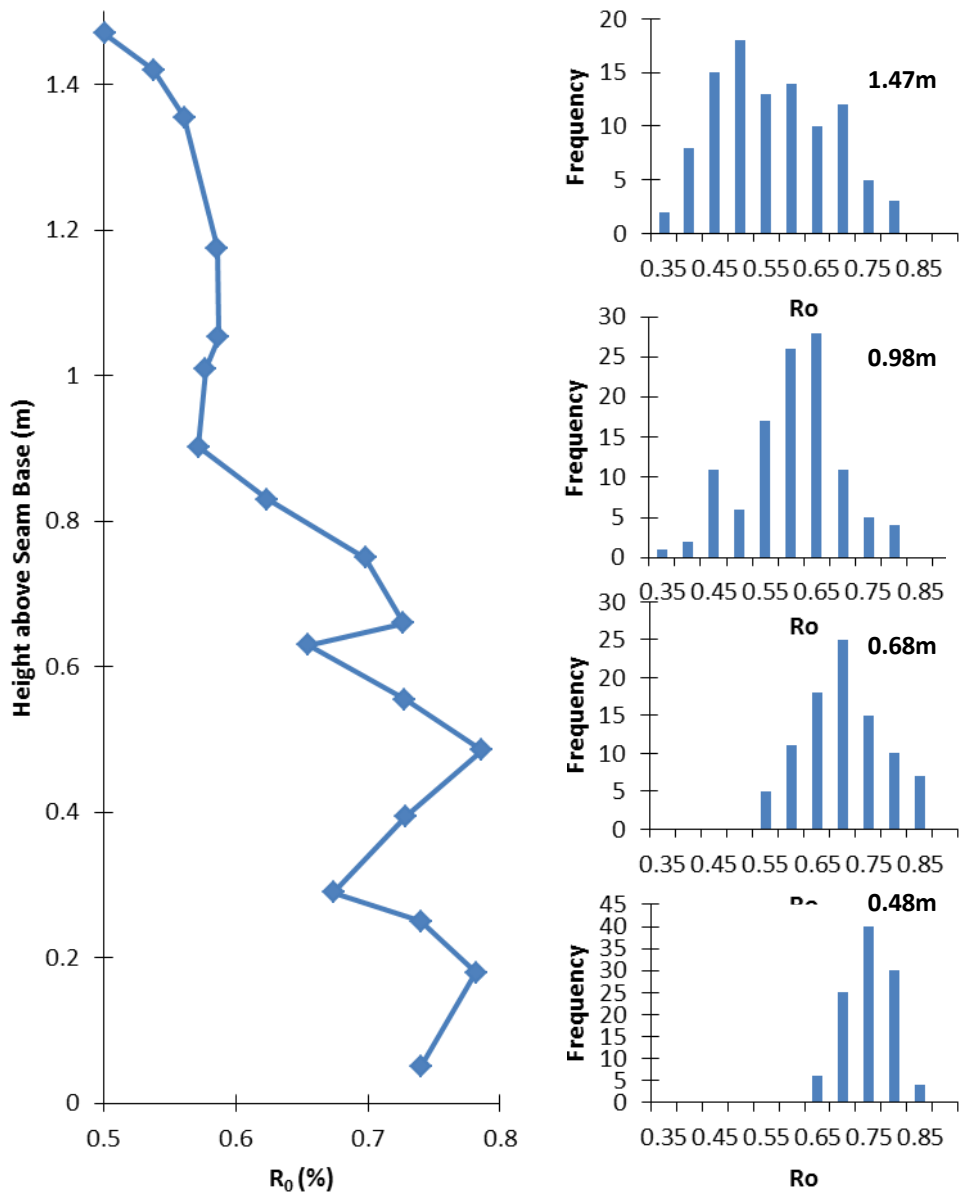
677 **Figure 3**

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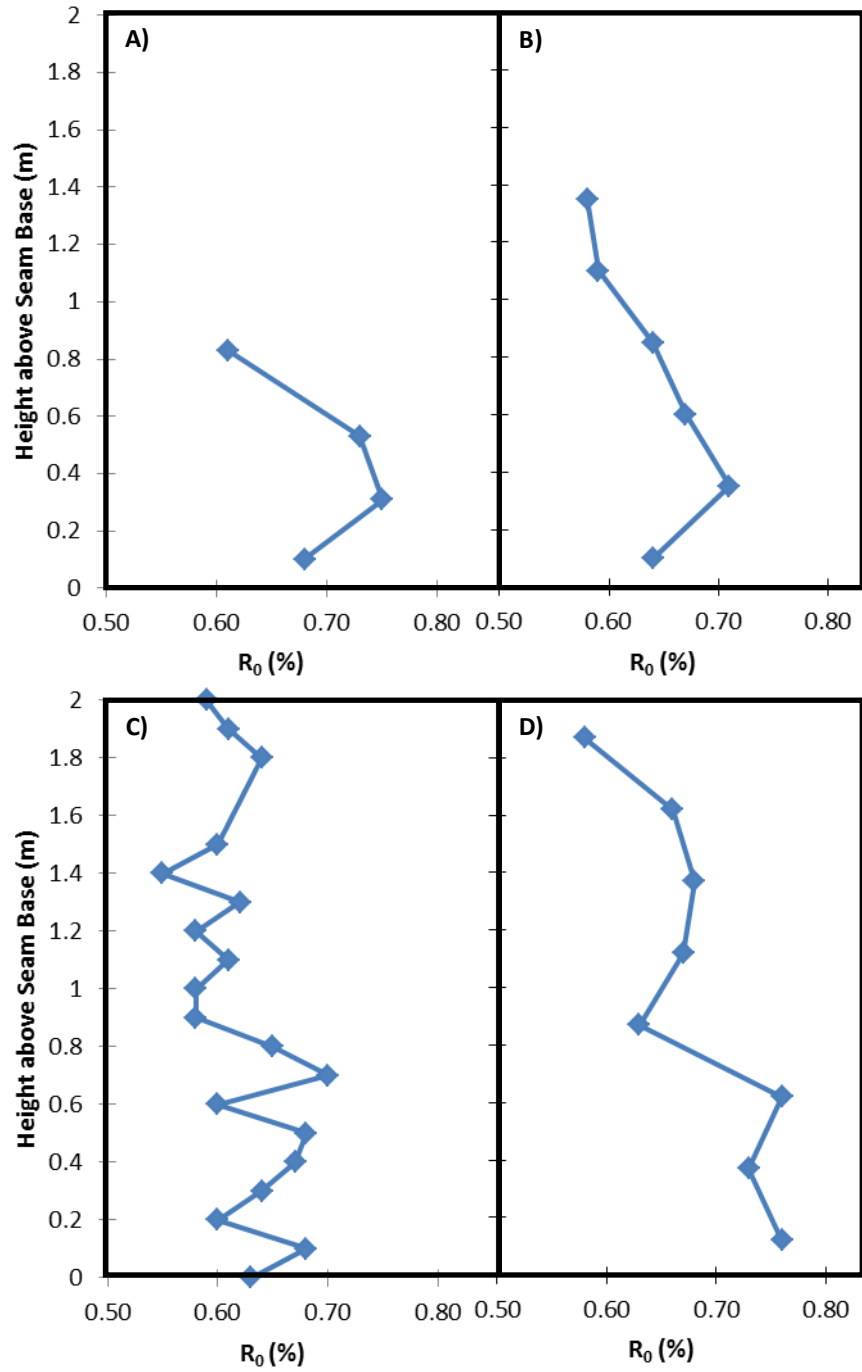
729 **Figure 4**

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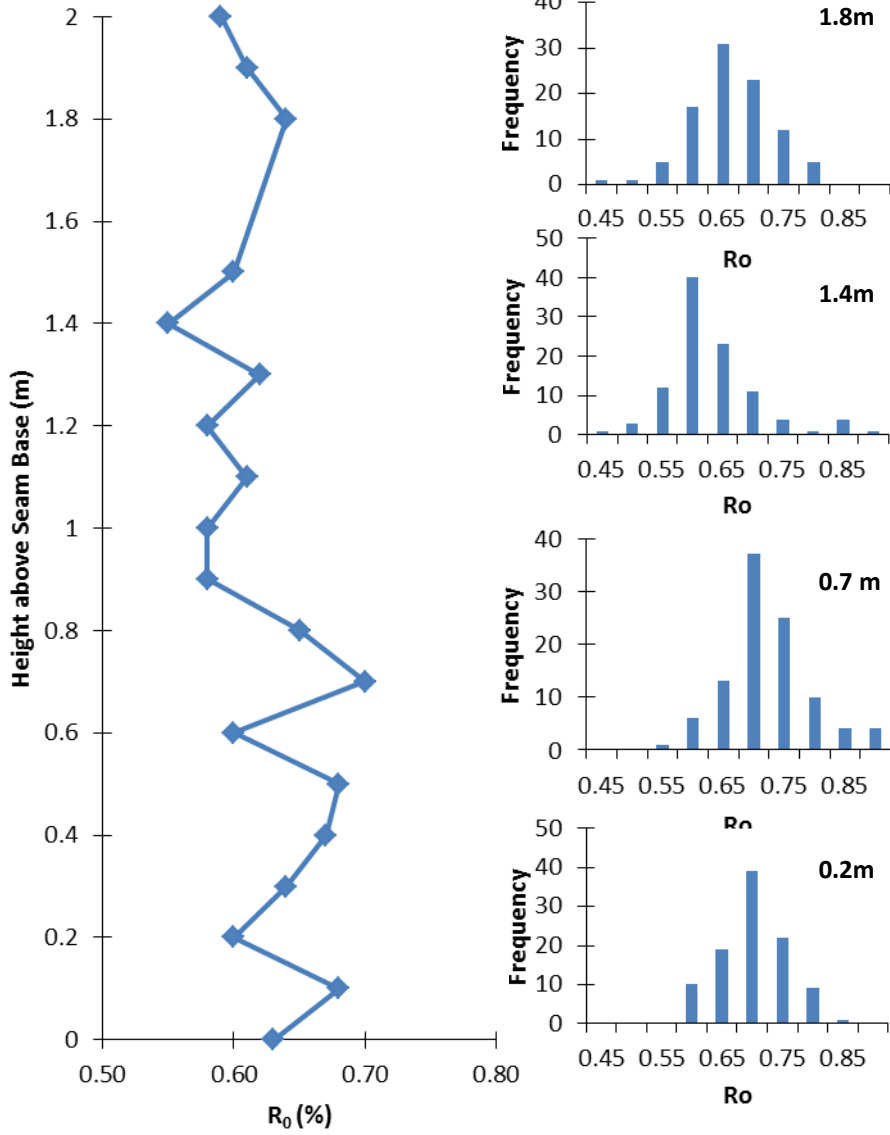
781 **Figure 5**

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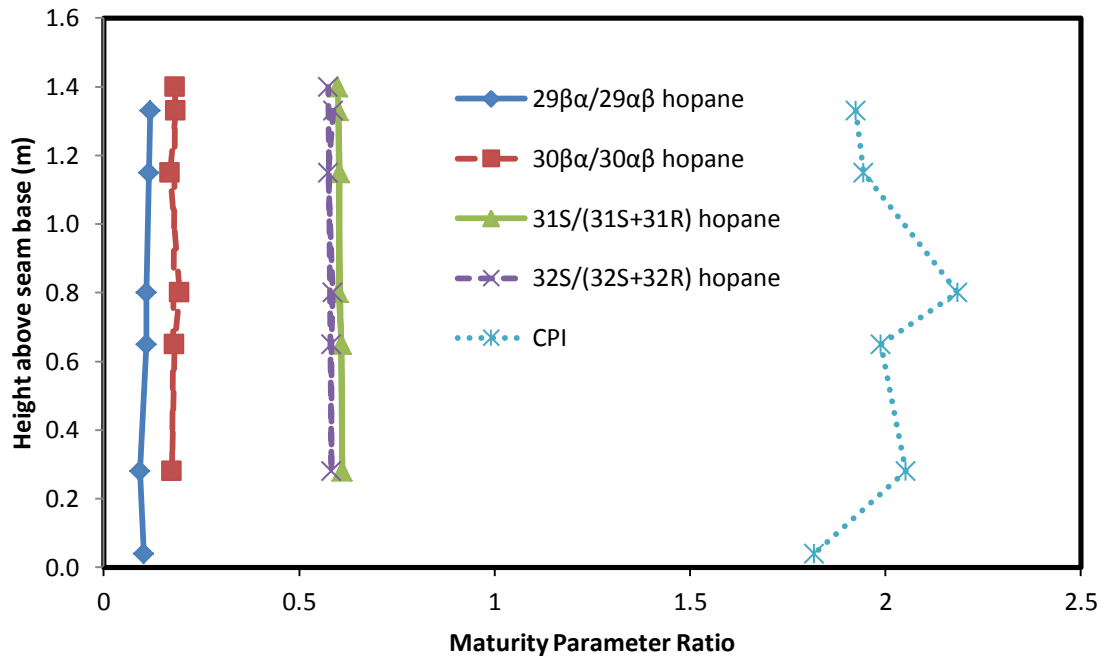


833 **Figure 6**

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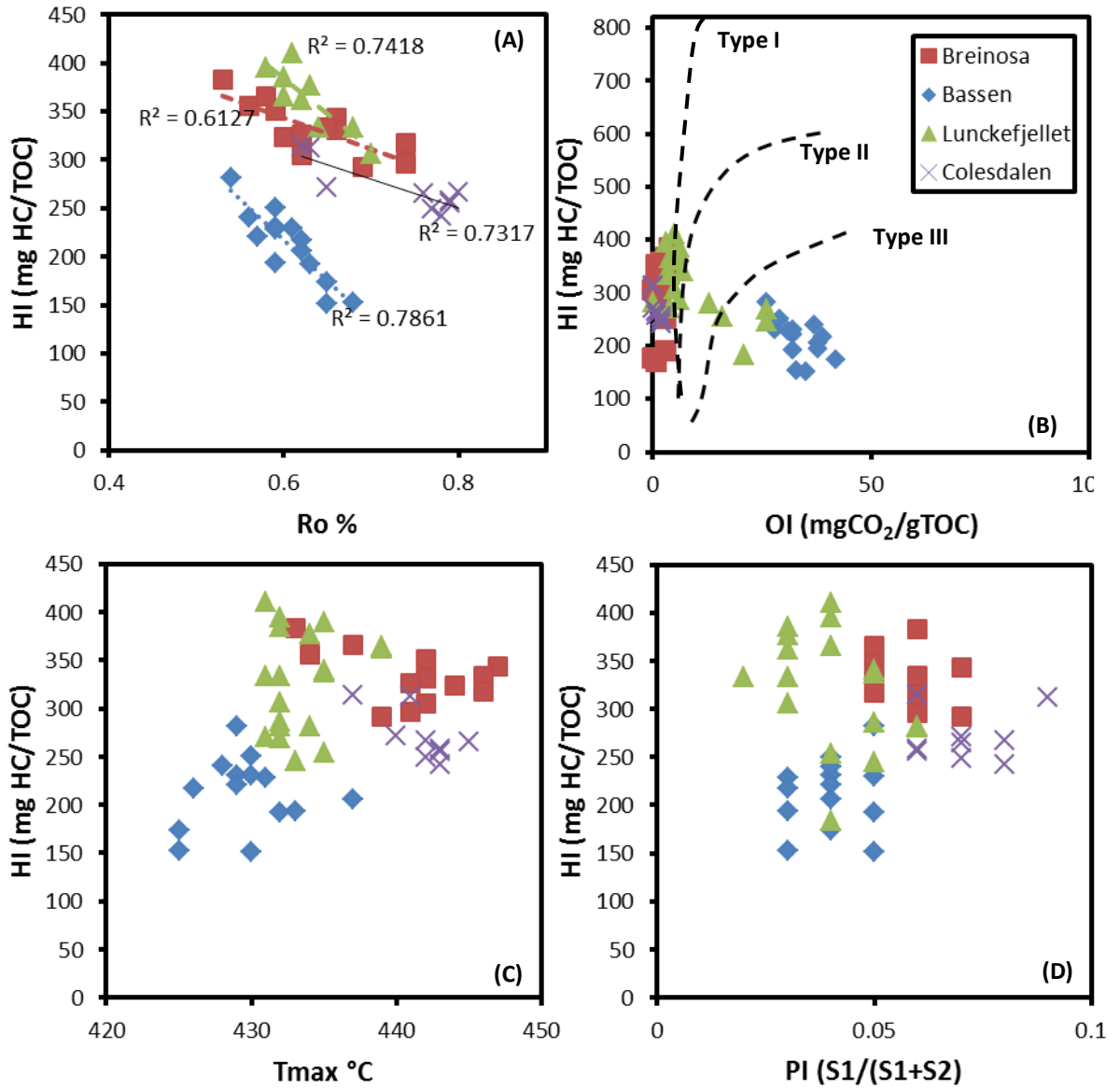
885 **Figure 7**  
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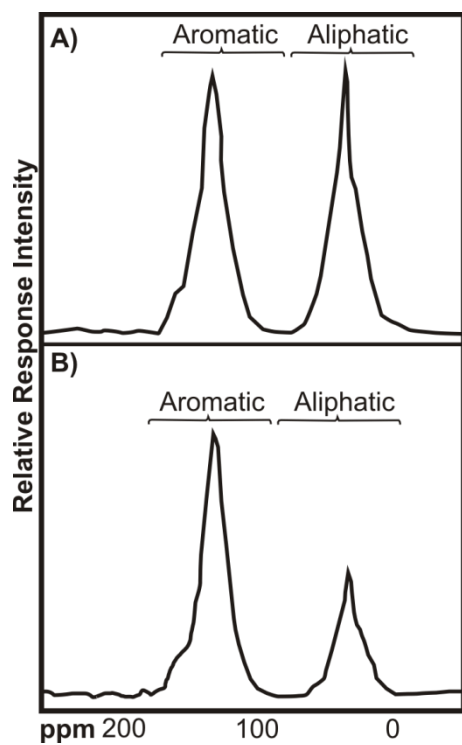
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Figure 8



971 **Figure 9**  
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1003**Table 1**

Seam	Seam Height (m)	R <sub>o</sub>	Pr/Ph	Pr/nC <sub>17</sub>	Ph/nC <sub>18</sub>	CPI(1)	Ts/Tm	C <sub>29</sub>	C <sub>30</sub>	C <sub>31</sub>	C <sub>32</sub>	C <sub>33</sub>	C <sub>29</sub> /C <sub>30</sub>	C <sub>35</sub> /C <sub>34</sub>	C <sub>29</sub>	C <sub>29</sub>	TS/TH	MPI- <sub>1</sub> <sup>a</sup>	Calculated R <sub>o</sub> <sup>b</sup>
			<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes	<i>n</i> -alkanes			
Svea Nord	Bulk	0.78	7.1	0.9	0.1	1.05	0.95	0.05	0.07	0.64	0.62	0.63	0.75	0.31	0.54	0.46	0.23	0.60	0.76
Longyear	Bulk	0.65	13.40	4.50	0.20	1.10	0.93	0.05	0.10	0.64	0.63	0.61	0.75	0.41	0.56	0.49	0.19	0.63	0.78
Svea Nord	0.07	0.77	4.48	24.55	4.43	1.46	0.93	0.09	0.08	0.58	0.58	0.58	0.58	0.43	0.42	0.51	0.21	0.48	0.69
Svea Nord	0.82	0.78	4.82	2.80	0.43	1.54	0.93	0.08	0.12	0.60	0.60	0.62	0.60	0.16	0.48	0.5	0.19	0.65	0.79
Breinosa Svea	0.75	0.78	3.51	22.48	3.77	1.41	0.94	0.07	0.09	0.59	0.59	0.62	0.63	0.23	0.51	0.51	0.19	0.51	0.71
Breinosa Svea	0.50	0.78	6.29	17.33	2.01	1.50	0.95	0.10	0.11	0.60	0.60	0.61	0.59	0.14	0.54	0.5	2	0.51	0.70
Breinosa Svea	1.00	0.79	3.53	2.70	0.66	1.54	0.93	0.08	0.09	0.62	0.62	0.64	0.56	-	0.47	0.53	0.2	0.49	0.69
Breinosa Svea	1.25	0.76	26.54	75.45	1.87	1.42	0.92	0.08	0.10	0.59	0.59	0.62	0.47	0.19	0.45	0.55	0.2	0.47	0.68
Todalen	-	0.75	20.99	119.83	2.93	1.39	0.93	0.06	0.07	0.60	0.60	0.63	0.55	0.28	0.47	0.49	0.21	0.57	0.74
Svarteper	0.73	0.65	3.01	0.68	0.27	1.40	0.91	0.10	0.05	0.58	0.58	0.59	0.50	-	0.48	0.52	0.2	0.76	0.86
Svarteper	0.94	0.65	6.49	9.99	0.97	1.57	0.95	0.09	0.13	0.60	0.60	0.61	0.61	0.16	0.49	0.51	0.21	0.56	0.74
Askeladden	0.33	0.64	2.95	6.23	1.73	1.39	0.94	0.11	0.10	0.59	0.59	0.62	0.54	0.29	0.48	0.5	0.21	0.52	0.71
Longyear	1.43	0.501	3.46	7.75	0.18	1.40	0.92	0.08	0.06	0.58	0.58	0.60	0.51	0.49	0.43	0.51	0.2	0.51	0.71
Longyear	1.43	0.538	11.44	12.22	1.34	1.59	0.93	0.11	0.07	0.60	0.59	0.60	0.67	0.30	0.45	0.49	0.18	0.51	0.71
Longyear	1.35	0.561	10.13	20.67	1.56	1.53	0.94	0.10	0.11	0.60	0.57	0.60	0.60	0.31	0.43	0.50	0.2	0.49	0.69
Longyear	1.17	0.586	11.41	17.37	1.15	1.56	0.95	0.09	0.12	0.60	0.59	0.62	0.62	0.30	0.45	0.50	0.21	0.55	0.73
Longyear	0.83	0.628	13.42	15.42	0.81	1.62	0.93	0.08	0.09	0.61	0.58	0.61	0.60	0.03	0.41	0.52	0.22	0.49	0.69
Longyear	0.66	0.655	12.11	5.29	0.33	1.69	0.93	0.08	0.10	0.61	0.58	0.60	0.62	0.41	0.41	0.52	0.23	0.55	0.73
Longyear	0.29	0.74	12.31	6.20	0.35	1.72	0.93	0.10	0.10	0.60	0.58	0.59	0.62	0.31	0.40	0.51	0.18	0.54	0.73
Longyear	0.07	0.74	13.60	2.34	0.16	1.69	0.94	0.11	0.12	0.60	0.57	0.61	0.63	0.31	0.42	0.51	0.19	0.45	0.67

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