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International Journal of Coal Geology 70 (2007) 277 – 291
www.elsevier.com/locate/ijcoalgeo

Thermal alteration of terrestrial palynomorphs in mid-Cretaceous organic-rich mudstones intruded by an igneous sill (Newfoundland Margin, ODP Hole 1276A)

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Received 12 March 2006; received in revised form 14 June 2006; accepted 25 June 2006
Available online 17 July 2006

Abstract

Most approaches used to reconstruct thermal alteration of sediments necessitate advanced, relatively expensive analytical techniques. We have evaluated the fidelity of a less costly, relatively simple approach of visually assessing sporomorph colours to determine thermal alteration. The sporomorph-based thermal alteration estimates were compared to vitrinite reflectance data from the same samples. As study material, we selected a succession of mid-Cretaceous (Albian) organic-rich clay- and siltstones intruded by a diabase sill that was recovered from Ocean Drilling Program (ODP) Hole 1276A, off Newfoundland.

Six sporomorph groups (SG), each consisting of morphologically well-defined, easily identifiable constituents with long stratigraphic ranges, were individually evaluated for their thermal alteration signals. These groups are: (1) leiotrilete spores of the genera *Biretisporites*, *Cyathidites*, *Deltoidospora*, *Dictyophyllidites*, *Gleicheniidites*, and *Leiotriletes* (SG-1; subdivided into three subgroups SG-1a, SG-1b and SG-1c with sporoderm thicknesses <1 µm, 1–1.5 µm and >1.5 µm, respectively); (2) trilete, rugulate spores of the genera *Camerozonosporites* and *Lycopodiacidites* (SG-2); (3) trilete, striate spores of the genera *Appendicisporites*, *Cicatricosisporites* and *Plicatella* (SG-3); and (4) the gymnosperm-pollen taxon *Classopollis torosus* (SG-4). Sporomorph colours were determined using Munsell colour standards under reproducible optical conditions. To minimize the potential influence of reworked specimens on the dataset, only the lightest 50% of all counted specimens per sporomorph group were evaluated for their thermal alteration signals.

The thermal alteration estimates from all sporomorph groups yield an internally consistent picture that is compatible with vitrinite reflectance data from the same samples. They indicate that downhole thermal alteration does not increase until 20 m above the igneous sill. A steep rise occurs only at 4.23 m above the sill, and thermal alteration peaks in the sample closest (2.17 m) to the sill. However, the different sporomorph groups exhibit varying degrees of fidelity with respect to deciphering thermal alteration. Factors influencing the precision of the thermal alteration signal include sporoderm thickness, character of surface ornamentation, resistance to reworking, and abundance in the sample material. Highest correlations with vitrinite reflectance data are observed for the thermal alteration values from SG-1b ($R=0.82$), SG-3 ($R=0.80$) and SG-4 ($R=0.80$). Hence, these groups are best suited for a sporomorph-based approach to reconstructing the thermal history of sediments. The highest correlation coefficient with vitrinite

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doi:10.1016/j.coal.2006.06.005

reflectance data is registered for SG-1b, the subgroup with the least variability of sporoderm thickness and the highest abundance in the sample material. This indicates that the study of morphologically similar, highly abundant specimens with strongly constrained sporoderm thickness variations yields the best results for the reconstruction of thermal alteration.

Keywords: Sporomorph; Spore colour; Maturity; Vitrinite reflectance; Albian; Ocean Drilling Program

1. Introduction

As temperature represents a key parameter in hydrocarbon generation, reconstructing the thermal history of sediments is a critical task in applied geosciences. Consequently, to determine thermal alteration, various optical and physicochemical methods have been developed. The most prominent optical methods are the measurements of vitrinite reflectance (e.g., Tissot and Welte, 1984; Teichmüller et al., 1998), organic matter fluorescence (e.g., Ottenjahn et al., 1974; van Gijzel, 1981) and the colour alteration in conodonts (e.g., Belka, 1990; Deaton et al., 1996). Physicochemical methods comprise the determination of T_{max} (e.g., Espitalié et al., 1985–87; Bordenave et al., 1993), the analysis of the temperature-dependent conversion of clay minerals from smectite to illite (e.g., Pytte and Reynolds, 1989; Pollastro, 1993), and the analysis of isomers of complex biomarkers (e.g., Mackenzie and McKenzie, 1983; Sosrowidjojo et al., 1996). With the exception of the conodont-based approach, these methods necessitate advanced, relatively expensive analytical techniques. The conodont-based approach, in turn, requires relatively large amounts of sample material, thus limiting its applicability to core samples, and can only be applied to open marine sediments from the Paleozoic and Triassic. In this context, the temperature-dependent colour change of sporomorphs (i.e., the pollen and spores of vascular plants) provides a promising alternative method for deciphering thermal alteration. With increasing maturity, the colour of sporomorphs changes from pale yellow to orange and brown to opaque. This colourimetric evolution is progressive, cumulative and irreversible (Correia, 1971; Yule et al., 1998). The analysis of sporomorphs to determine thermal alteration has the advantage that only a few grams of sample material and a standard light microscope are required. Sporomorphs are widely distributed in terrestrial and marine sediments since the first appearance of spore-producing plants in the Ordovician and their rapid proliferation in the Silurian (e.g., Richardson, 1996; Steemans, 1999; Stricanne et al., 2006). They are especially abundant in organic-rich, fine-grained sediments that represent oil-prone source rocks (e.g., Traverse, 1988; Tyson, 1995). The fact that the measurements are made on organic particles that are an integral part of the hydrocarbon-sourcing kerogen rather than on potentially unrelated particles from bulk samples highlights the relevance of sporomorph colour analysis for hydrocarbon exploration (e.g., Grayson, 1975; Marshall, 1991). Moreover, sporomorph colour variations are particularly sensitive in the lower range of thermal maturation where they can yield more precise information than vitrinite reflectance values (Marshall, 1991; Jeans et al., 2005). Finally, the palynological preparations can be directly used for palynostratigraphic age determinations and for obtaining paleoenvironmental information.

Although sporomorph colours have been utilized in paleothermal estimates for more than 70 years (e.g., Stadnichenko, 1929), this approach has remained less prominent than other methods such as vitrinite reflectance. This is because consistency in sporomorph-based estimates of thermal maturation can be compromised by the dependence of colour variations on variations in wall thickness and composition as well as by the potential presence of reworked specimens (Batten, 1996). Nevertheless, sporomorph colour changes have been successfully utilized in many studies of thermal maturity (see Gray and Boucout, 1975; Smith, 1983; Marshall, 1990; and Batten, 1996, for reviews). The available approaches are either qualitative or quantitative. Qualitative approaches are based on the visual assessment of sporomorph colours according to more or less well-defined scales and provide an especially rapid and inexpensive method of assessing kerogen maturity. Ideally, these scales consist of a standard of colour slides or standardized colour charts. The fact that various colouration scales are available and most laboratories use in-house scales and standards has led to a relatively poor comparability of the results. Commonly used qualitative scales include (1) the Etat de Conservation index of Correia (1967, 1971) and Correia and Peniguel (1975), a 1–6 scale for different palynofacies constituents including sporomorphs, dinoflagellate cysts, acritarchs, chitinozoans, and plant debris; (2) the Thermal Alteration Index (TAI) of Staplin (1969, 1982), ranging from 1 to 5 and based on spores, cuticles, and amorphous sapropelic debris; (3) the Spore Colouration Index of Robertson

Research Group (Haseldonckx, 1979; Barnard et al., 1980), ranging from 1 to 10; and (4) the Spore Colouration Index of Batten (1980, 1982), ranging from 1 to 7. To increase the consistency and reproducibility of spore colouration data, Pearson (1982, 1984) published a colour chart based on ten defined colours with Munsell reference numbers that he related to the TAI scale of Staplin (1969, 1982).

In an effort to provide quantitative scales comparable in precision and reproducibility to that of vitrinite reflectance, quantitative approaches based on the measurement of the translucency of sporomorphs using a photometric unit, have been developed. They were pioneered by Gutjahr (1966) who linked the translucency of sporomorphs to coal rank. Later studies include those of Grayson (1975), Smith (1983), Lo (1988), Lerche and McKenna (1991), Marshall (1991), and Yule et al. (1998, 1999). However, while quantitative approaches are less subjective than qualitative approaches, they also require relatively complex and expensive analytical techniques. Thus, a significant advantage of spore colouration analysis over other methods that determine thermal alteration is lost. It is therefore not surprising that several workers have again investigated the robustness of not fully quantitative approaches to determine sporomorph colours. Collins (1990) showed that the simple visual assessment of sporomorph colours based on a comparison to a standard of colour slides yields remarkably consistent results. The high fidelity of sporomorph colour records with regard to thermal alteration has recently been stressed by Kalkreuth et al. (2002). When evaluating different optical and chemical maturity parameters as measured on Jurassic material from the North Sea, these authors found a higher correlation coefficient between estimated spore colour indices and measured vitrinite reflectance values than between measured T_{max} and vitrinite reflectance values.

In view of the above, this paper explores a systematic, semiquantitative approach for determining thermal maturation based on the visual assessment of sporomorph colours. Special emphasis was given to analyze morphologically and taxonomically well-defined sporomorph groups to avoid the problem of colour variations in dependence of variable sporoderm thicknesses and/or varying chemical compositions. To maximize the applicability of our approach, we focussed on sporomorph groups with long stratigraphic ranges. Sporomorph colours were determined using Munsell colour standards under reproducible optical conditions. To determine the fidelity of the dataset, the colour estimates from each group were compared to vitrinite reflectance data from the same samples. As study material, we have selected a mid-Cretaceous (Albian) succession of organic-rich clay- and siltstones intruded by an igneous sill that was recovered from Ocean Drilling Program (ODP) Hole 1276A, off Newfoundland. This succession provides an ideal natural laboratory for our purposes as it is characterized by high sedimentation rates, relatively uniform lithology, and a strong thermal maturity gradient due to the postdepositional emplacement of a diabase sill (Shipboard Scientific Party, 2004). This provides the opportunity to examine pronounced colour change over a relatively short time interval, thus minimizing evolutionary and/or environmentally driven changes in the sporomorph associations and yielding an internally consistent dataset. In addition, this analysis can constrain the thermal influence of the diabase sill on the overlying mid-Cretaceous sediments and the thickness of sediments overlying the sill at the time it was emplaced.

2. Study site

The studied section comprises a 220-m-thick interval of mid-Cretaceous sediments that was drilled in ODP Hole 1276A, off Newfoundland (Shipboard Scientific Party, 2004; Fig. 1). The motivation for drilling at Site 1276 was to better understand the syn- and post-rift history of the Newfoundland–Iberia conjugate margins. While basement was not reached at this site, 936.9 m of early Oligocene to mid-Cretaceous sediments were cored, reaching a total depth of 1736.9 mbsf, an estimated 100–200 m above basement according to seismic data (Shipboard Scientific Party, 2004).

The examined section reaches from 1395 to 1611 mbsf. Lithologically, it is predominantly comprised of dark gray mudrocks with minor sand- to silt-based turbidites and black shales. At 1612.11 mbsf, metasediments consisting of hydrothermally altered mudstone and calcareous sandstone were encountered. Further downhole, the top of a diabase sill was reached at 1612.83 mbsf (Shipboard Scientific Party, 2004). Based on nannoplankton and dinoflagellate cyst biostratigraphic data, the section has an early to middle Albian age (Shipboard Scientific Party, 2004). The age of the sill has been estimated to be 82.5–105.9 Ma at Site 1276 (Karner and Shillington, 2005) and has since been radiometrically dated at 105.95 ± 1.78 Ma and 104.7 ± 1.7 Ma (S. Hart, 2005, personal communication).

3. Material and methods

3.1. Palynology

Twenty-four samples from the mid-Cretaceous section of ODP Hole 1276A were investigated for the colour

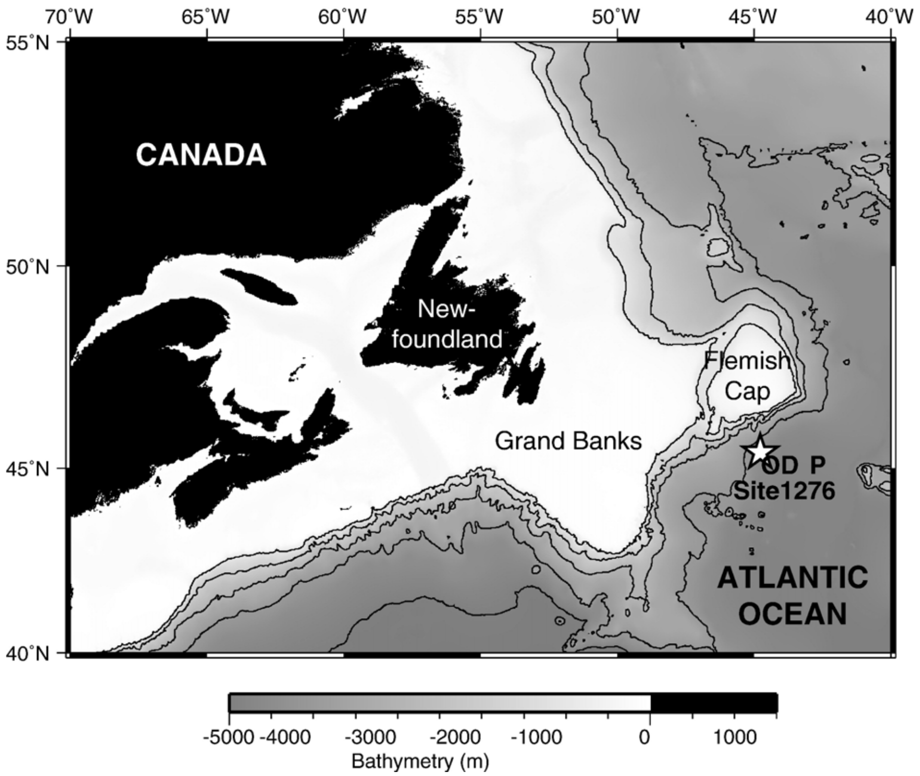


Fig. 1. Location of ODP Site 1276A. Map showing gridded bathymetry extracted from the GEBCO Digital Atlas (IOC, IHO, BODC, 2003). Contour interval is 1000 m. Site 1276A is indicated with a white star.

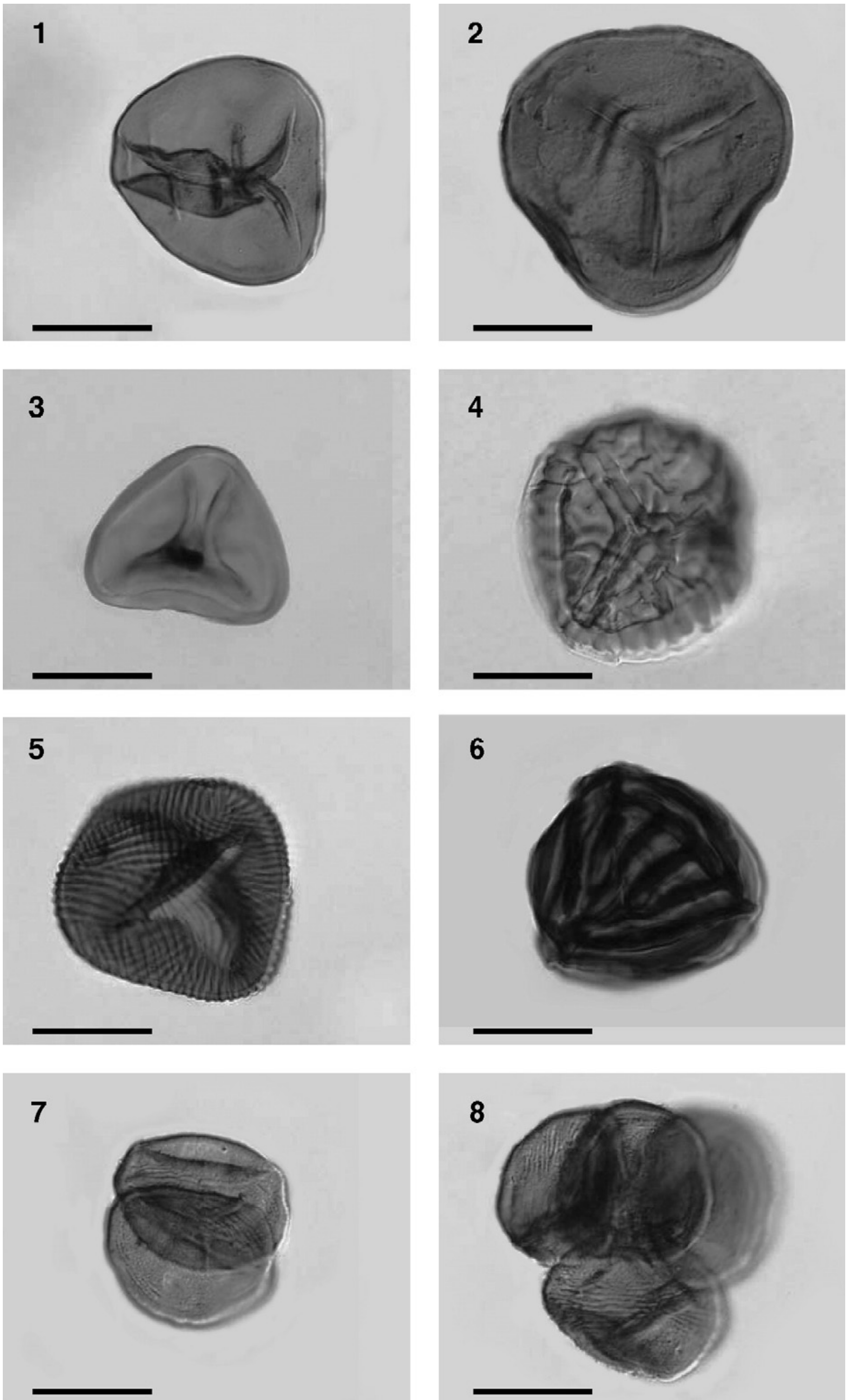
changes recorded in sporomorphs. Samples were processed following standard palynological techniques (e.g., Pross, 2001). After careful cleaning, 10–15 g of sediment were treated with 33% HCl to dissolve carbonates and with 40% HF to dissolve silicates. Subsequent to each chemical preparation step, the residues were sieved through a 6 μm nylon mesh using distilled water. To avoid processing-induced bleaching of the sporomorphs, no oxidation was performed. After extensive mixing to obtain homogeneity, strew mounts were prepared using glycerine jelly as a mounting medium. All materials are filed in the collection of the Institute of Geosciences, University of Frankfurt, Germany.

Following a first palynological survey of the samples, four sporomorph groups, each comprised of morphologically closely related taxa, were defined. Further criteria for the definition of sporomorph groups were that their representatives are stratigraphically long ranging and occurred in sufficient numbers and with high consistency in the samples. Sporomorph Group 1 (SG-1) consists of leiotrilete spores of the genera *Biretisporites*, *Cyathidites*, *Deltoidospora*, *Dictyophyllidites*, *Gleicheniidites*, and *Leiotriletes*. This group was further subdivided based on the sporoderm thicknesses of the encountered specimens. Sporoderm thicknesses were estimated using an eyepiece micrometer. Sporomorph

Fig. 2. Representatives of the sporomorph groups investigated for thermal alteration signals. Scale bars equal 20 μm.

1. *Cyathidites* sp. (Sporomorph Group 1-a). Sample 1082.05 mbsf
2. *Cyathidites* sp. (Sporomorph Group 1-b). Sample 1529.85 mbsf
3. *Deltoidospora* sp. (Sporomorph Group 1-c). Sample 1082.05 mbsf
4. *Camerozonosporites* sp. (Sporomorph Group 2). Sample 1082.05 mbsf
5. *Cicatricosisporites* sp. (Sporomorph Group 3). Sample 1295.06 mbsf
6. *Cicatricosisporites* sp. (Sporomorph Group 3). Sample 1128.55 mbsf
7. *Classopollis torosus* (Sporomorph Group 4). Sample 1082.05 mbsf
8. *Classopollis torosus* (Sporomorph Group 4). Sample 1529.85 mbsf.

Subgroup 1a (SG-1a) comprises specimens with sporoderm thicknesses <1 μm, Sporomorph Subgroup 1b (SG-1b) comprises specimens with sporoderm thicknesses of 1 to 1.5 μm, and Sporomorph Subgroup 1c (SG-1c) comprises specimens with sporoderm thicknesses >1.5 μm. Sporomorph Group 2 (SG-2) was



defined to comprise trilete, rugulate spores of the genera *Camerozonosporites* and *Lycopodiacidites*. Sporomorph Group 3 (SG-3) consists of trilete, striate spores, comprising specimens of the genera *Appendicisporites*, *Cicatricosisporites* and *Plicatella*. Sporomorph Group 4 (SG-4) consists of the gymnosperm-pollen taxon *Classopollis torosus* and is thus monospecific. Representatives of each sporomorph group are illustrated in Fig. 2.

From each sample, three strew mounts were investigated under transmitted light for representatives of the sporomorph groups defined above using an Olympus BX 50 light microscope. To warrant reproducibility of the results, colour determinations were carried out under constant optical conditions using a magnification of 630×. Illumination was from a 9 V tungsten bulb with a colour temperature of 3200 K; no filters were applied.

Sporomorph colours were estimated on the sporoderms of unfolded, well-preserved specimens based on a visual comparison to Munsell colour standards as suggested by Pearson (1984). Following the work of Staplin (1969, 1982), the utilized thermal alteration scale reaches from 1 to 5, with low values reflecting low thermal alteration and vice versa. To increase the resolution of the scale, intermediate values of 0.3 and 0.7 between full values were used. The thermal alteration scale used in this study and the respective colour standards for each TAI value as depicted in Munsell colour charts (GretagMacbeth LLC, 617 Little Britain Road, New Windsor, NY 12553) are presented in Table 1. To reduce the influence of potentially reworked specimens on the obtained TAI data, only the lightest 50% of all counted specimens per sporomorph group and sample were used as a basis for further evaluations. This procedure is based on the consideration that the lightest colour recorded in a sporomorph group from a sample reflects the maximum thermal influence the sample was exposed to. The numbers of specimens counted from each sporomorph group and further used as a basis for the calculation of TAI values are presented in Table 2.

Table 1
Calibration of sporomorph thermal alteration index (TAI) utilised in this study to Munsell colour standards

TAI	Sporomorph colour	Munsell prod. no.	Hue/Value/Chroma
1	Pale yellow	17,391	7.5Y 9/4
1.3	Yellow	20,520	7.5Y 9/8
1.7	Sunny yellow	19,638	5Y 8.5/12
2	Yellow orange	14,253	2.5Y 8/12
2.3	Bright orange	13,800	10YR 7/12
2.7	Orange	12,424	10YR 6/10
3	Yellowish brown	15,816	10YR 5/6
3.3	Dark yellowish brown	17,209	10YR 4/4
3.7	Very dark grayish brown	15,814A	10YR 3/2
4	Very dark gray	19,365	10YR 2.5/1
5	Black (opaque)	n/a	N1

Low TAI values reflect low thermal alteration and vice versa. The calibration is based on colour estimates on the sporoderms of unfolded, well-preserved sporomorphs under defined optical conditions. See Section 3.1 for further discussion.

Table 2
Number of specimens from each sporomorph group as used for the calculation of TAI values, and vitrinite reflectance data from each sample studied

Depth (mbsf)	SG-1a	SG-1b	SG-1c	SG-2	SG-3	SG-4	Vitrinite reflectance (%)
1395.06	1	15	3	14	3	5	0.48
1421.72	5	5	1	0	2	1	0.63
1444.71	6	6	3	1	2	3	0.61
1466.44	7	10	5	1	3	1	0.66
1482.19	11	7	4	8	4	8	0.61
1498.92	1	9	1	1	1	5	0.64
1515.37	3	20	4	1	7	2	0.62
1529.85	12	24	16	4	10	8	0.6
1543.45	6	18	6	1	14	3	0.55
1556.17	14	21	7	2	14	9	0.51
1565.16	9	5	1	3	9	0	0.65
1575.5	4	7	3	2	6	7	0.62
1586.54	5	6	3	1	11	5	0.61
1588.95	7	9	7	1	14	4	0.58
1592.61	7	5	7	3	9	3	0.63
1599.26	2	16	12	2	14	9	0.66
1600.85	3	15	12	3	14	9	0.68
1601.97	8	8	1	0	5	1	0.68
1604.98	5	20	4	1	12	12	0.66
1606.43	1	13	1	3	12	8	0.68
1607.2	7	6	1	1	8	1	0.72
1609.14	4	8	1	2	8	4	0.78
1610.25	5	14	4	0	10	2	0.75
1610.66	7	6	1	0	5	3	0.87

3.2. Vitrinite reflectance

Vitrinite reflectance was determined from polished particulate mounts for all palynologically sampled horizons. After crushing each sample to a grain size of ≤3 mm, representative amounts of pellets were impregnated with epoxy resin using a vacuum impregnation device. To ensure sample homogeneity, final mounts were prepared from 5-mm-thick slices cut vertically into the middle part of the pellet blocks. The slices were re-embedded in epoxy resin at the bottom of a mould. Polishing followed the procedures for carbonaceous material. As most samples turned out to be sensitive to water, samples were ground under dry conditions, and polishing was carried out using an alcohol-based lubricant.

Microscopic analyses of the polished particulate mounts were carried out with a Leitz DMRX-MPVSP microscope photometer in both incident white-light and UV+violet-light illumination (fluorescence mode) using oil immersion objectives (magnification: 200–1000×). Random reflectance measurements on vitrinite were performed following procedures outlined by the International Handbook of Coal Petrography (ICCP, 1963, 1971, 1975, 1993, 1998). One hundred vitrinite readings were recorded for each sample. In addition, the qualifying system for reflectance analysis that is currently developed and tested by an ICCP working group has been applied (Borrego et al., 2004). In each sample, the rank–vitrinite population was identified, and the resulting mean random reflectance (mean value of all accepted rank–vitrinite readings) was calculated. This value was used for comparison with palynomorph colours to determine thermal alteration levels (see Table 2 for vitrinite reflectance data from each sample).

4. Results

4.1. Palynology

All samples yielded well-preserved, diverse palynofloras consisting predominantly of sporomorphs. Other palynomorphs such as dinoflagellate cysts and acritarchs occur in subordinate numbers.

4.1.1. Sporomorph Group 1 (SG-1)

The three subgroups of SG-1, which consist of leiotrilete spores of the genera *Biretisporites*, *Cyathidites*, *Deltoidospora*, *Dictyophyllidites*, *Gleicheniidites*, and Leiotriletes separated based on sporoderm thicknesses, are present in all samples investigated. Sporomorph Group 1a, comprising specimens with sporoderm thicknesses <1 μm, has been encountered with average counts of 11 specimens (minimum: one specimen, 1395.06 mbsf; maximum: 28 specimens, 1556.17 mbsf) per sample. In the interval from 1395.06 to 1592.61 mbsf, TAI values show a relatively irregular pattern with an overall trend towards higher values at greater depths (Fig. 2a). Below 1592.61 mbsf, TAI values increase rapidly. Lower TAI values are again recorded in the depth interval between 1607.2 and 1609.14 mbsf. Further downhole, TAI values increase again, reaching a maximum of 2.43 at 1610.66 mbsf, which is situated closest to the igneous sill (Fig. 3a).

Sporomorph Group 1b, characterized by sporoderm thicknesses between 1 and 1.5 μm, occurs with mean counts of 23 specimens per sample (minimum: nine specimens, 1592.61 mbsf; maximum: 48 specimens, 1529.85 mbsf) and is thus the most abundant sporomorph group within the sample series. TAI values from SG-1b remain relatively stable in the upper part of the examined section. From 1565.16 to 1604.98 mbsf, they increase slightly, but show a rather irregular pattern. Below 1606.43 mbsf, in close proximity to the sill, a strong increase in TAI to values as high as 2.83 in the lowermost sample is recorded (Fig. 3a).

Sporomorph Group 1c, characterized by sporoderm thicknesses >1.5 μm, is represented by nine specimens per sample on average (minimum: one specimen, 1606.43 mbsf; maximum: 32 specimens, 1529.85 mbsf). TAI values from SG-1c exhibit a relatively irregular pattern in the upper part of the section. From 1592.61 mbsf downhole, a strong TAI increase is observed. After a transient return to lower values at 1606.43 mbsf, TAI values reach a maximum of 3.0 in the lowermost sample (Fig. 3a).

Summarizing, the TAI records from all SG-1 subgroups reveal a consistent picture. In the upper part of the section, a trend of only slightly increasing TAI values is recorded. This trend is, however, obscured by a relatively irregular pattern in the data especially from subgroups SG-1a and SG-1b. Downhole from 1592.61 mbsf, TAI values increase markedly in all subgroups. After a transient drop centered around 1606.43 mbsf, maximum TAI values are reached in the lowermost sample, which is situated closest to the igneous sill.

4.1.2. Sporomorph Group 2 (SG-2)

Sporomorphs attributed to SG-2, i.e., trilete spores of the genera *Camerozonosporites* and *Lycopodiacidites*, are relatively rare within the sample series. Total counts never exceed 28 specimens per sample and most samples yielded five specimens or less. The TAI curve for SG-2 exhibits the most irregular pattern of all sporomorph groups. One single sporomorph found at 1444.71 mbsf shows an erratic TAI value of 3.00. Other than that, the highest TAI values are reached closest to the sill (Fig. 3b).

4.1.3. Sporomorph Group 3 (SG-3)

Sporomorphs assigned to SG-3, consisting of striate trilete spores of the genera *Appendicisporites*, *Cicatricosisporites* and *Plicatella*, were found in all samples, with a mean value of 16 specimens per sample (minimum: three specimens, 1601.97 mbsf; maximum: 28 specimens, 1556.17 mbsf). From 1395.06 to 1592.61 mbsf, no significant TAI changes are recognised (Fig. 3b). Similar to SG-2, an outlier marked by a TAI value of 2.67 occurs at 1444.71 mbsf; this value is, however, only based on two specimens. Downhole from 1599.26 mbsf, TAI values

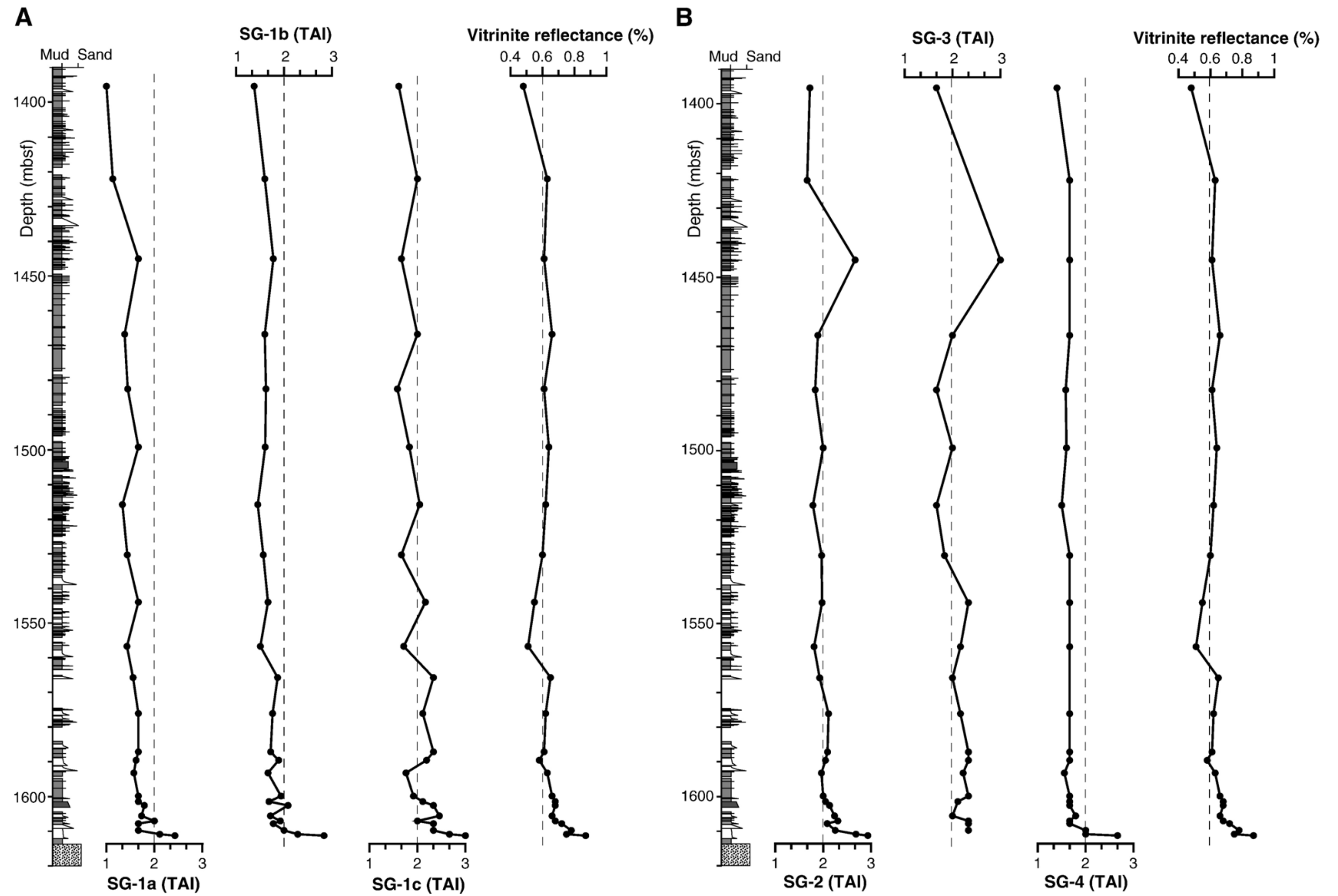


Fig. 3. (a) Thermal alteration indices of sporomorph groups SG-1a, SG-1b and SG-1c (left) and vitrinite reflectance data (%*R*; right) from the mid-Cretaceous succession drilled in ODP Hole 1276A. Lithology after Shipboard Scientific Party (2004). See Sections 4.1 and 4.2 for discussion. (b) Thermal alteration indices of sporomorph groups SG-2, SG-3 and SG-4 (left) and vitrinite reflectance data (%*R*; right) from the mid-Cretaceous succession drilled in ODP Hole 1276A. Lithology after Shipboard Scientific Party (2004). See Sections 4.1 and 4.2 for discussion.

increase strongly. After a transient reduction at 1606.43 mbsf, the maximum TAI recorded is reached at 1610.66 mbsf, closest to the igneous sill (Fig. 3b).

4.1.4. Sporomorph Group 4 (SG-4)

Classopollis torosus, the only constituent of SG-4, was found in all samples, except at 1565.16 mbsf, with a mean of 10 counts per sample (minimum: one specimen, 1421.72 mbsf; maximum: 24 specimens, 1604.98 mbsf). The TAI values estimated for SG-4 yield the most consistent curve of all examined sporomorph groups (Fig. 3b). Similar to the TAI records from the other sporomorph groups, the highest TAI value is measured for the sample adjacent to the igneous sill.

4.2. Vitrinite reflectance

Vitrinite reflectance at the top of the studied section (1395.06 mbsf) is 0.48%. From 1395.06 to 1590 mbsf, it primarily alternates between 0.6% and 0.66%. A slightly reduced reflectance value is recorded at 1556.17 m (Fig. 3). Downhole from 1590 mbsf, a rapid increase of reflectance values takes place. The maximum reflectance (0.87%) is recorded at 1610.66 mbsf closest to the igneous sill.

5. Discussion

5.1. Consistency of TAI signals from different sporomorph groups

The TAI estimates from all sporomorph groups investigated yield similar trends and hence provide an internally consistent picture of thermal alteration (Fig. 3a, b). For the samples investigated, representatives of sporomorph subgroup SG-1a tend to yield slightly lower TAI values than representatives of subgroup SG-1b and especially SG-1c. Highest TAI values tend to occur in representatives of sporomorph groups SG-2 and SG-3. This pattern indicates that within sporomorph group SG-1 representatives of subgroup SG-1a are primarily slightly lighter than representatives of the subgroups SG-1b and especially SG-1c. In turn, representatives of sporomorph groups SG-2 and SG-3 are primarily slightly darker than representatives of sporomorph group SG-1.

From the top of the section to around 1590 mbsf, thermal alteration remains relatively low. Although obscured through a relatively irregular pattern in the data especially from subgroups SG-1c, SG-2, and SG-3, there is a slight trend towards increased values at greater depth. Closer to the igneous sill, from 1592.61 mbsf downhole, strong TAI changes are registered for all sporomorph groups, indicating strongly increased thermal alteration. However, all sporomorph groups exhibit relatively low TAI values from 1606.43 to 1607.2 mbsf, which suggests reduced thermal alteration in that interval. Maximum thermal alteration as derived from TAI estimates occurred at 1610.66 mbsf, 2.17 m above the igneous sill (Fig. 3a, b).

Varying irregularities in the TAI signals suggest different fidelities of the studied sporomorph groups as thermal alteration indicators. Hence, to assess the fidelity of the studied sporomorph groups in recording thermal alteration, TAI values from each group were tested for their correlation with vitrinite reflectance data from the same samples (Fig. 4).

Overall, there are relatively few irregularities in the TAI data from all three subgroups of SG-1. TAI values from any given sample are consistently lower for sporomorphs assigned to SG-1a (comprising specimens with sporoderm thicknesses <1 μm) than for sporomorphs assigned to SG-1b and SG-1c (comprising specimens with sporoderm thicknesses of 1–1.5 μm and >1.5 μm, respectively, Fig. 3a, b). Sporoderm thickness thus exerts a strong control on sporomorph colour, with thicker sporoderms thicknesses appearing darker than thinner sporoderms and vice versa.

Subgroup 1b yields the most consistent signal with the least irregularities (Fig. 3). Stronger irregularities occur in the TAI records from SG-1a and SG-1c. These observations are in accordance with the correlation coefficients calculated for SG-1a, SG-1b, and SG-1c with vitrinite reflectance data. The highest correlation with vitrinite reflectance data emerges for SG-1b with a correlation coefficient of $R=0.82$ (Fig. 4). Similar, but slightly lower correlation coefficients ($R=0.74$) are calculated for subgroups SG-1a and SG-1c. The reduced fidelity of SG-1a and SG-1c in comparison to SG-1b is ascribed to the fact that both these groups are comprised of specimens with a higher variance in sporoderm thickness than SG-1b. Therefore, a greater scatter in TAI values is likely.

To obtain the highest possible homogeneity of TAI estimates, a narrow delineation of the sporoderm thicknesses to be measured is mandatory, yet there should be sufficient specimens to warrant credibility. Because SG-1b has yielded the highest counts of all sporomorph groups investigated despite its narrow definition, this subgroup appears to be particularly well suited for determining thermal alteration. Additionally, the morphologically simple genera *Biretisporites*, *Cyathidites*, *Deltoidospora*, *Dictyophyllidites*, *Gleicheniidites*, and *Leiotriletes* are easily identified, thus offering the potential to be evaluated even by non-experts.

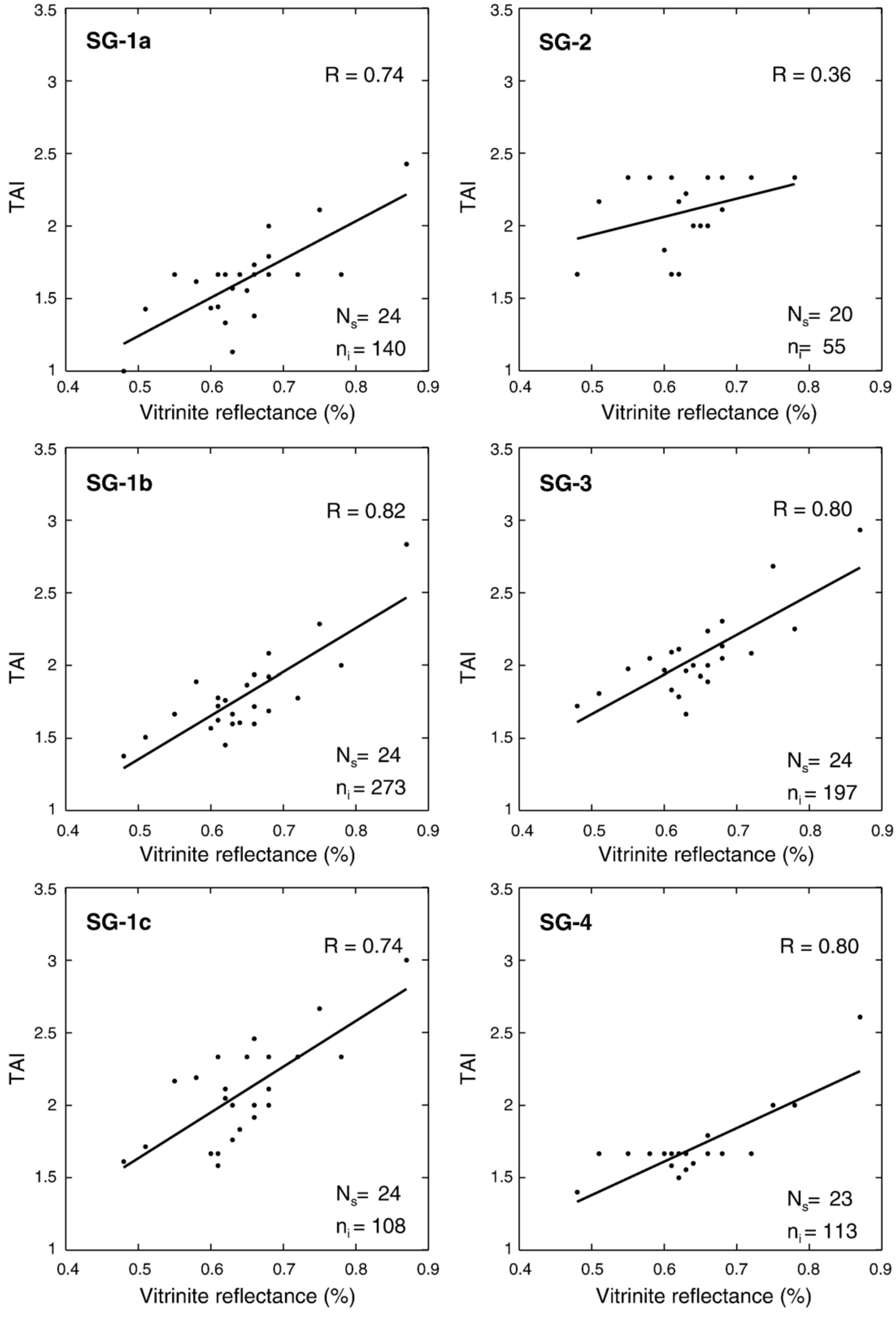


Fig. 4. Cross-plots of thermal alteration indices of investigated sporomorph groups against vitrinite reflectance data (%*R*) from the same samples. n_s = number of samples; n_i = total number of measured sporomorphs.

Although the TAI trends are similar to those from the other sporomorph groups, there is a significant variability in the TAI data from SG-2 (Fig. 3). The strong TAI variability in this irregularly ornamented sporomorph group is also reflected in the lowest correlation coefficient with vitrinite reflectance data ($R=0.36$; Fig. 4) of all sporomorph groups investigated. Outliers in the TAI record from SG-2 are most obvious where data points are based on single specimens such as at 1444.71 mbsf, where a TAI value of 3.0 was measured. Since neither the TAI values from any other of the sporomorph groups investigated nor vitrinite reflectance data are consistent with such high thermal alteration, this value is most probably due to reworking. This is in accordance with the lithology of the studied section, consisting predominantly of dark gray mudrocks with minor sand- to silt-based turbidites (Shipboard Scientific Party, 2004). It can be argued that members of SG-2 are generally more robust to oxidation and/or redeposition than the relatively fragile sporomorphs with thin sporoderms and no ornamentation as they comprise the sporomorph groups SG-1 and SG-4. Hence, they can be expected to occur relatively more often in turbidite layers where other, less robust sporomorphs have not been preserved. Therefore the probability of measuring reworked specimens is higher for SG-2 than for SG-1 and SG-4. Together with the fact that SG-2 is by far the least abundant sporomorph group in the examined section and irregularly ornamented sporomorphs generally tend to yield a high scatter in TAI measurements due to the thickness variations in their sporoderms (cf. Gutjahr, 1966), SG-2 appears to be of only limited value in deciphering thermal alteration.

Sporomorph group 3, which consists of striate, regularly ornamented trilete spores of the genera *Appendicisporites*, *Cicatricosisporites* and *Plicatella*, yields a TAI curve that is both consistent with curves derived from the other studied sporomorph groups and with the vitrinite reflectance record (Fig. 3). The high fidelity of SG-3 in recording thermal alteration is further documented by the high correlation coefficient ($R=0.80$) with vitrinite reflectance data (Fig. 4). At first glance, the high fidelity in reflecting thermal alteration seems surprising considering that representatives of SG-3 seem highly resistant to reworking due to the presence of up to 4 μm thick striae. In analogy to SG-2, this should lead to a rather irregular TAI record. However, representatives of SG-3 are more than three times more abundant in the sample material than those of SG-2, and in contrast to those from SG-2, they are present in all samples investigated. Therefore it can be assumed that the presence of reworked specimens had relatively little impact on the calculated TAI values due to the much higher abundance of allochthonous specimens as compared to SG-2. Moreover, the ornamentation in the genera *Appendicisporites*, *Cicatricosisporites* and *Plicatella* consists of parallel striations and is extremely regular, thus allowing an internally much more consistent measurement of TAI values for this group than for SG-2.

Sporomorph group 4, which consists exclusively of *Classopollis torosus* pollen grains and is thus monospecific, yields the most uniform curve of thermal alteration when compared to the TAI records from the other sporomorph groups and the vitrinite reflectance curve (Fig. 3). The TAI data obtained from SG-4 yield the second highest correlation coefficient with the vitrinite reflectance data ($R=0.80$) of all sporomorph groups investigated (Fig. 4). These observations, together with the fact that *Classopollis torosus* is an easily recognizable, highly abundant element within Mesozoic sporomorph assemblages, suggest that SG-4 is very well suited for the determination of thermal alteration.

Summarizing the above, the TAI signals from the sporomorph groups investigated are internally consistent, although there is a strong scatter in the TAI signals especially from SG-2. A comparison with vitrinite reflectance data from the same samples yields the highest correlation coefficients and thus the highest fidelity in reflecting thermal alteration for SG-1b ($R=0.82$), SG-4 ($R=0.80$), and SG-3 ($R=0.80$). Representatives of all these groups are easily recognized. Their high abundance in the sample material in combination with our approach of using only the lightest 50% of all counted specimens per sporomorph group as a basis for TAI calculations significantly reduces the potential influence of reworked specimens on the obtained TAI records. The subdivision of SG-1 in subgroups based on sporoderm thickness yielded the highest correlation coefficient with vitrinite reflectance data for SG-1b, the subgroup with the strongest constraint in sporoderm thickness variability and the highest abundance in the sample material. This suggests that the approach of studying highly abundant, morphologically similar specimens with strongly constrained sporoderm thickness variations can be advantageous to studying monospecific or -generic, but less abundant groups.

The correlation coefficients of TAI values with vitrinite reflectance data obtained in our study are slightly lower than the values of Kalkreuth et al. (2002). Based on Jurassic material from the North Sea, these authors achieved a correlation coefficient between spore colour and vitrinite reflectance of $R=0.90$. This correlation appears particularly high considering the fact that Kalkreuth et al. (2002) did not distinguish between indigenous (i.e., lighter) and reworked (i.e., potentially darker) spores. Hence, it seems reasonable to assume that the material of Kalkreuth et al. (2002) contained less reworked sporomorphs than the succession from the mid-Cretaceous off Newfoundland, a scenario that is well supported by the very different lithologies and sedimentary settings studied. Kalkreuth et al. (2002) performed their studies on clays, shales and coals from an epicontinental basin, whereas the sediments studied in the present contribution are derived from the foot of a continental slope, with a substantial proportion of the sediments having been deposited by turbidity currents (Shipboard Scientific Party, 2004; Hiscott, in press).

5.2. Implications for the thermal history of mid-Cretaceous sediments at Site 1276A

Contrary to an expected downhole increase in thermal alteration, neither vitrinite reflectance nor the most correlative TAI data (i.e., SG-1b and SG-3; Fig. 3) show a clear trend towards greater maturity throughout most of the studied section (1395–1601 mbsf). Any such trend, if present, must have been masked by the natural variability of organic particles (both sporomorphs and vitrinite). This variability may result from different degrees of reworking and preservation of organic particles, as well as from maturation-induced changes due to variations of the mineral matrix (Espitalié et al., 1980, 1984) or vitrinite reflectance suppression due to variable alginite content (e.g., Price and Barker, 1985).

It is only within a distance of about 11 m from the sill that an unambiguous gradient appears. Vitrinite reflectance increases from 0.66% to 0.87% over a distance of 4.23 m (Fig. 3). Similarly, TAI values from all sporomorph groups exhibit a significant rise over the same interval. This gradient signals the overprint due to sill emplacement. Our data suggest that the near-field thermal effect of the igneous intrusion was stronger than the background thermal alteration related to burial heating.

Vitrinite reflectance values between 0.5% and 0.7% are generally considered to correlate with the upper limit of the oil window (onset of major liquid hydrocarbon generation; Dow, 1977; Teichmüller et al., 1998). Corresponding TAI values between 1.35 and 1.95 for SG-1b, between 1.66 and 2.21 for SG-3, and between 1.38 and 1.84 for SG-4 as obtained in this study (compare Fig. 4) also mark the upper limit of the oil window. Thus, most of the studied section is marginally mature with respect to oil generation. However, because the dominant kerogen type in this interval is humic or type III kerogen (Pletsch et al., submitted for publication), such that oil generation was negligible in spite of favourable thermal conditions.

In the lowermost part of the studied section (1610.66 mbsf) and beneath (Pletsch et al., submitted for publication), vitrinite reflectance reaches 0.87%, just above the conventional definition for the onset of major thermal gas generation at 0.8% (Dow, 1977; Teichmüller et al., 1998). Liquid hydrocarbons detected in this interval and closer to the sill testify to the activity of this spatially limited source kitchen.

Summarizing, the combined evidence provided by TAI and vitrinite reflectance data has allows valuable insights into the thermal overprint of the mid-Cretaceous section. These two independent organic maturity indicators revealed the lack of a trend for most of the studied section, whereas indications for a significant thermal alteration are restricted to a narrow (present thickness 4.23 m) interval (the aureole) above the igneous sill. It is reasonable to assume that the thickness of the aureole has been significantly reduced by compaction since the intrusion event. Assuming an intrusion depth between 50 and 550 m of sediment and using the age–depth relationship of Karner and Shillington (2005) results in original aureole thicknesses ranging from 6.5 to 13.5 m. Thus, the original thickness of the aureole was likely around 10 m, similar to the thickness of the sill.

In a study of igneous intrusions into wet sediments of the Australian Gippsland Basin, Barker et al. (1998) explained similar, narrow profiles of decreasing vitrinite reflectance and other thermal indicators to indicate the incipient formation of convection cells that allowed for a particularly rapid cooling of the intrusive heat source through the advection of cool pore water. A similar incipient convection scenario is proposed to explain the rapid decrease of Thermal Alteration Indices and vitrinite reflectance values at only a few meters above the intrusive contact (Pletsch et al., submitted for publication).

6. Conclusions

Based on a succession of mid-Cretaceous sediments intruded by a igneous sill that was drilled in Ocean Drilling Program Hole 1276A (Newfoundland Margin), thermal maturation has been semiquantitatively estimated through the visual comparison of sporomorph colours to Munsell colour standards under defined optical conditions. To maximize the applicability of our approach, special attention was paid to studying stratigraphically long-ranging sporomorph taxa. The problem of colour variations in dependence of sporoderm thicknesses was addressed by analyzing morphologically and taxonomically well-constrained sporomorph groups. The fidelity of the thermal alteration data derived from these sporomorph groups was examined by testing their correlation with vitrinite reflectance data from the same samples. The most salient results of our study are as follows:

(1) The semiquantitative estimate of sporomorph colours using Munsell colour standards under defined, constant optical conditions yields internally consistent, reproducible results. The measurement of different morphologically and taxonomically defined sporomorph groups indicates that different sporomorph groups yield varying degrees of fidelity with respect to deciphering thermal alteration. Factors influencing the fidelity of the thermal alteration signal include sporoderm thickness, character of the surface ornamentation, resistance to reworking, and abundance in the sample material.
(2) Of the four sporomorph groups investigated, Sporomorph Group 1 (SG-1; consisting of *Biretisporites*, *Cyathidites*, *Deltoidospora*, *Dictyophyllidites*, *Gleicheniidites*, and *Leiotriletes*), Sporomorph Group 4 (SG-4; *Classopollis torosus*), and Sporomorph Group 3 (SG-3; *Appendicisporites*, *Cicatricosisporites* and *Plicatella*) yielded the clearest thermal alteration data. The TAI records from all three groups exhibit the highest correlation coefficients with vitrinite reflectance data. A partition of SG-1 into three subgroups based on sporoderm thickness showed that the subgroup with the narrowest range in sporoderm thickness yielded the highest correlation with vitrinite reflectance data.
(3) Thick-walled, irregularly ornamented spores as represented by Sporomorph Group 2 (SG-2; *Camerozonosporites* and *Lycopodiacidites*) yielded very irregular thermal alteration data that show only a low correlation with vitrinite reflectance values. Their strongly variable sporoderm thicknesses render it difficult to obtain internally consistent data. Moreover, they are by far the least abundant sporomorph group investigated. This, in combination with their high robustness to chemical and physical degradation, can potentially cause an overrepresentation of reworked specimens in the assemblages evaluated, thus yielding potentially misleading estimates of thermal alteration.
(4) Thermal Alteration Indices and vitrinite reflectance values do not reveal a significant downhole trend for most of the studied section. It is only at 4.23 m above the igneous sill that both thermal indicators raise steeply. They reach a maximum closest to the sill. This trend reflects the spatially limited maturation effect of the igneous intrusion. Advection of cool pore fluids through a relatively open pore network may have played a role in the cooling history of this igneous body.

Acknowledgments

We thank P.A. Hochuli, J.E.A. Marshall and B. van de Schootbrugge for suggestions. U. Kotthoff provided technical support. The constructive reviews of an anonymous reviewer and of International Journal of Coal Geology Editor in Chief J.C. Hower are highly appreciated. This research used samples and data provided by the Ocean Drilling Program. The Ocean Drilling Program is sponsored by the participating countries, including the German Research Council, whose support is gratefully acknowledged. Funding for this research was provided by the Federal Institute for Geosciences and Natural Resources president's grant BL00-L04009, and by USSAC.

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