Palaeo-thermal and coalification history of Permo-Carboniferous sedimentary basins of Central and Western Bohemia, Czech Republic: first insights from apatite fission track analysis and vitrinite reflectance modelling

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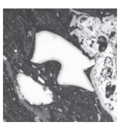

Apatite fission track analysis (AFTA) and vitrinite reflectance (VR) modelling were integrated to reveal palaeo-thermal evolution of the Upper Carboniferous coal-bearing basins of Central and Western Bohemia. Thermal modelling using AFTA has shown that the Carboniferous sediments were subjected to maximum temperatures ranging from ~75 °C in the SW to more than ~120 °C in the NE, which generally concluded during pre-Triassic times. Thermal records show a gradual cooling between ~280–180 Ma, followed by a long Mesozoic to Cenozoic Period of thermal stability, during which time, the sediments resided at constant temperatures of ~50–75 °C. From ~30 Ma onwards, accelerated cooling and erosion occurred, resulting in the present-day exposure of rocks on the surface. The EASY%R_o modelling of average VR values that range from 0.59% R_r in the SW to 0.77% R_r in the NE, predicted maximum palaeo-temperatures ranging from ~85 °C to ~135 °C, respectively; these were attained soon after sediment deposition. A coalification grade of about 0.40–0.50% R_r characteristic of sub-bituminous coals, was already completed during the late Carboniferous and/or early Permian times. Later post-Permian heating did not have any substantial impact on the maturation of Carboniferous organic matter. By combining VR thermal modelling with local stratigraphic information we provide evidence that the coalification process occurred very rapidly. A coalification grade of ~0.40% R_r was attained during 2–4 m.y. after peat deposition, or even earlier. In addition, abundant sub-bituminous coal clasts embedded within the Carboniferous strata suggest that coalification proceeded close to the surface. These data collectively imply extremely high thermal gradients (around or above 200 °C/km) that must have prevailed during the Permo-Carboniferous thermal climax. The syn-sedimentary volcanic and/or igneous activity combined with effects of heat advection due to hot fluids circulating through the strata could have accounted for this dramatic geothermal setting, which had vanished by the end of the Palaeozoic Era. • Key words: coalification, organic matter diagenesis, geothermal gradient, heat flow evolution, Variscan orogeny, Bohemian Massif.

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During the last decade, a number of studies have applied the technique of apatite fission track (AFT) analysis (AFTA) to various geological formations in order to reconstruct their low-temperature thermo-chronologies, exhumation rates and/or landscape geomorphological evolution (see also Lisker *et al*. 2009, Green & Duddy 2012 and Enkelmann & Garver 2016 for reviews on AFTA applications to geology). With respect to the Bohemian Massif, most of these studies have concentrated on its peripheral zones, where the AFTA technique documented an intensive Mesozoic denudation (*e.g*. Hejl *et al*. 1997, 2003; Thomson & Zeh 2000; Ventura & Lisker 2003;

Aramowicz *et al*. 2006; Danišík *et al*. 2010; Sobczyk *et al*. 2015). On the other hand, thermal studies of rocks forming the interior parts of the Bohemian Massif that yielded substantially older AFT ages (*e.g.* Vamvaka *et al*. 2013) have been limited so far to the Lower Palaeozoic and Neoproterozoic sediments of the Barrandian area in the central part of the Czech Republic (Glasmacher *et al*. 2002; Suchy *et al*. 2002; Filip & Suchý 2004; Suchý *et al*. 2007, 2015).

In order to bridge this gap and to expand our knowledge of the low-temperature history of younger sedimentary formations from interior parts of the Bohemian Massif, we examined the apatite fission-track thermo-chronology of the Upper Carboniferous sediments that overlie Neoproterozoic and Lower Palaeozoic basement rocks in Central and Western Bohemia. In an attempt to achieve a deeper insight into the thermal history of these deposits, we combined the AFT technique with independent vitrinite reflectance kinetic modelling. The combination of these two techniques permitted the reconstruction of the palaeo-temperature record, extending deeper into the thermal past of the rocks, beyond the limits of the AFT technique alone.

Our data showed that the Carboniferous succession was subjected to an intense, but short-lived Variscan thermal event. An important by-product of our research was new evidence on the duration of the coalification process, which proceeded rapidly at a shallow burial level, being accelerated by highly elevated Variscan heat flows.

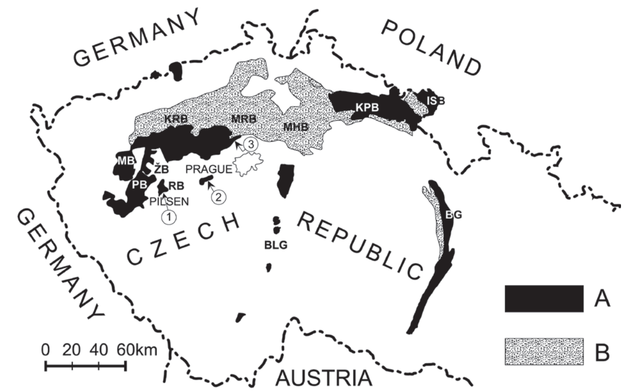

Figure 1. Schematic map showing the occurrence of Carboniferous terrestrial deposits and principal Carboniferous sedimentary basins in the Czech Republic (modified after Mísař *et al*. 1983); A – surface outcrops of Carboniferous deposits; B – Carboniferous deposits under the cover of younger sedimentary units. Abbreviations of Carboniferous basins: BG – Boskovice Graben; BLG – Blanice Graben; ISB – Intra-Sudetic Basin; KPB – Krkonoše Piedmont Basin; KRB – Kladno-Rakovník Basin; MB – Manětín Basin; MHB – Mnichovo Hradiště Basin; MRB – Mšeno-Roudnice Basin; PB – Plzeň Basin; RB – Radnice Basin; ŽB – Žihle Basin. Positions of the localities examined for their palaeo-thermal histories are also shown: 1 – Radnice-Ovčín; 2 – Nižbor (Stradonice, Lísek); 3 – Lobeč.

Geological setting and previous studies

A series of Late Palaeozoic coal-bearing basins – notably Plzeň, Manětín, Žihle, Kladno-Rakovník and Mšeno-Roudnice basins – occur in the central and western part of the Czech Republic, covering an area of about 6,000 km^2 (Pešek *et al*. 1998; Fig. 1). These extensional depressions, filled with terrestrial Permo-Carboniferous deposits, developed in the interior of Variscan Europe, some 300–400 km to the south from the North Variscan Foredeep (Ziegler 1990, McCann *et al*. 2006). The tectonic nature of these basins was variously reported. Havlena (1982) suggested a model of an extensive asymmetric mega-graben that subsided into the Neoproterozoic crystalline basement, but according to later studies, the basins originated as wrench-fault – activated depressions triggered by oblique convergence of Gondwana and Baltica plates (Pašek & Urban 1990).

In most basins the deposition began almost synchronously during the late Westphalian (near the Duckmantian–Bolsovian boundary) and lasted until the early Permian (Cisuralian) time. The depth of sedimentary fill preserved in individual basins varied from several tens of metres to about 1,440 m, depending on local variations in depositional history, subsidence rate, and the intensity of post-Carboniferous erosion. Sedimentation was often interrupted by numerous hiatuses and diastems, controlled by various tectonic and climatic events (*i.e.* Opluštil *et al.* 2016, Martínek *et al.* 2017). The sedimentary section is commonly subdivided into four lithostratigraphic formations, some of which are further separated into individual members. The two oldest formations (*i.e.* Kladno and Týnec formations) were dominated by fluvial sediments, while the lacustrine sediments were more widespread in the younger units (*i.e.* Slaný and Líně formations). The sandstones and claystones of the Kladno and Slaný formations were mostly grey and contained coal seams, while the two other units were mostly red-coloured and barren (Fig. 2). These sediments were deposited in variable continental settings ranging from extensive alluvial flood plains and incised river valleys to swamps and ephemeral lakes (Pešek 1996, 2001; Opluštil 2005; Lojka *et al.* 2016). The main sedimentary input into the basins was derived from local sources exposed in nearby upland areas, including some deeply eroded Variscan granitic intrusions and low-grade metamorphosed Neoproterozoic to Lower Palaeozoic rocks (Kukal 1983; Pešek 1996, 2004; Žák *et al.* 2018).

The coal rank of the Carboniferous coals increased with depth in many basins (Havlena 1964, Skoček 1976) where it ranged from ~0.40 to 1.11% R_r (Pešek 2001). Regional variations in coal rank between individual basins have been documented for coal seams of the Radnice Member, which range from 0.57–0.68% R_r in the West (the Plzeň Basin) up to 0.9–1.0% R_r in the East (the Kladno-Rakovník Basin), corresponding to the rank of bituminous coals (Pešek 1996, 2001). More detailed regional coalification studies have been carried out in the Plzeň Basin, where economically important coal seams were extensively mined in the past (Malán 1985, Pašek 1988). Substantially less coalified organic matter-rich Cretaceous (Cenomanian) sediments locally overlaid the Carboniferous strata. This structural relationship provides evidence that the main stage of coalification culminated before Cenomanian times (Havlena 1964).

In contrast to the stratigraphy and subsurface architecture of Carboniferous basins, which have been studied in detail, their thermal history, palaeo-heat flow regime and post-Variscan evolution have seldom been addressed. Šafanda *et al*. (1990) were the first to use vitrinite reflectance data from several deep boreholes, along with Buntebarth's (1982) calculation method in an attempt to estimate Carboniferous–Permian thermal gradients in these basins. More recently, Holub *et al.* (1997) applied the technique of time-temperature modelling of vitrinite reflectance and biomarker reactions to reveal the thermal history of the Mšeno-Roudnice Basin. Nevertheless, substantial uncertainties still persist about the original thickness, timing and magnitude of uplift, and the amount of erosion of the Carboniferous and Permian strata (see also Franců *et al*. 1998, Glasmacher *et al.* 2002, Filip & Suchý 2004, Pešek & Martínek 2012 and the discussions therein).

In the present study, we examine palaeo-thermal exposure and the time-temperature evolution of the Kladno Formation, which is the oldest stratigraphic unit of the Carboniferous succession. These strata are exposed on the surface at many places along the southern erosional margin of the basins in Central and Western Bohemia (Fig. 1).

The Kladno Formation, which comprises mostly grey, 360–415 m thick deposits, consists of two lithostratigraphic units – the Radnice Member and the Nýřany Member separated by a major depositional hiatus (Fig. 2). Sediments of the Radnice Member were deposited over eroded Lower Palaeozoic or Neoproterozoic basement rocks, during a period of significant tectonic activity. This is evidenced by abrupt facial changes and the variable thickness of Radnice sedimentary strata, which range from several metres to 279 m. The dominant lithologies are arkoses and aleuropelites, but conglomerates, volcanic tuffs, and economically important bituminous coal seams also occur locally.

The Nýřany Member accumulated after a 3.6 m.y. long depositional break (Opluštil *et al.* 2016) that is reflected in a prominent disconformity between its basal sediments and

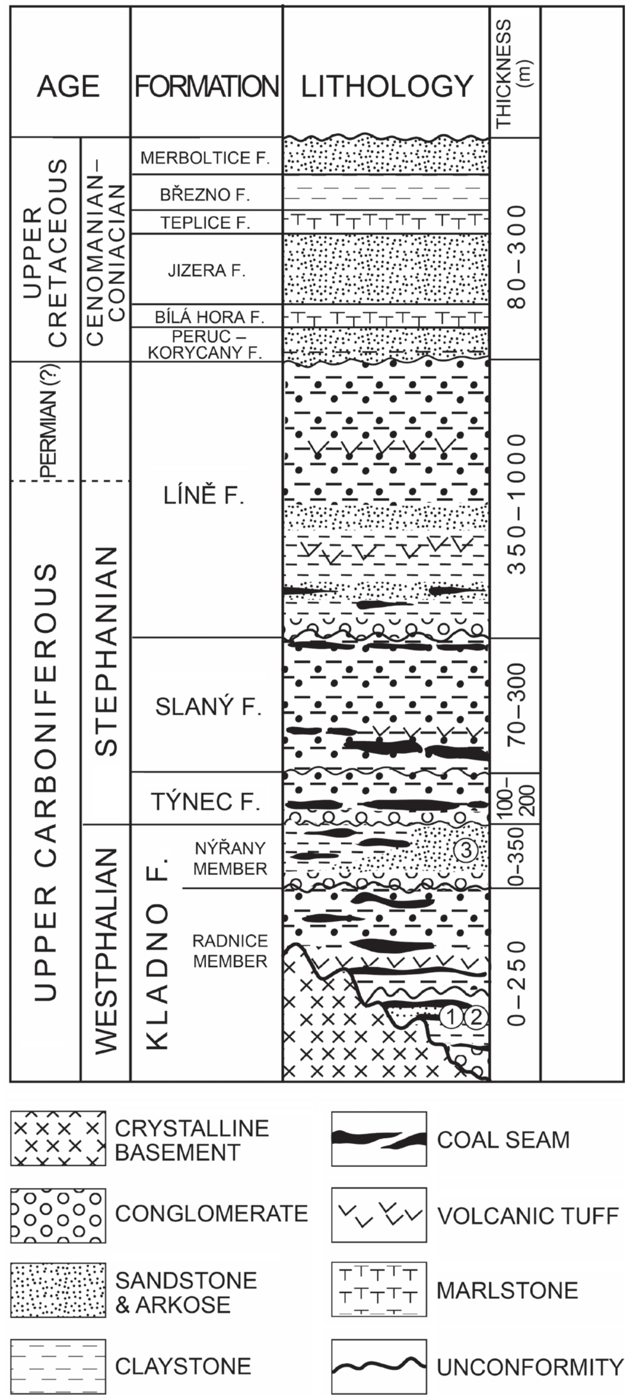

Figure 2. Schematic lithostratigraphy of Permo-Carboniferous (Holub 1982) and Cretaceous deposits for the central part of the Czech Republic (Čech *et al*. 1980). Stratigraphic positions of the samples are also plotted. Abbreviations: 1 – Radnice-Ovčín; 2 – Nižbor (Stradonice, Lísek); 3 – Lobeč; F. – Formation. See the text for details.

the underlying Radnice member. The average thickness of the Nýřany Member in the Kladno-Rakovník Basin was about 335 m. The lowermost part of the unit consisted of conglomerates interbedded with arkoses and aleuropelites that graded upward into arkoses and aleuropelites locally intercalated with layers of volcanic tuffs and thin bituminous coal seams (see Pešek 2004 and Pešek & Sivek 2016 for the details on local stratigraphy and lithology).

Sample details

The thermal exposure of Carboniferous sediments was investigated at three surface outcrops where the sediments yielded sufficient amounts of apatite grains for fission track analyses and allowed for simultaneous assessment of coaly fragments for respective vitrinite reflectance values. The absolute ages of the samples were calibrated toward the new chronostratigraphic timetable for Central and Western Bohemian Carboniferous basins (Opluštil *et al*. 2016) as 314 Ma for Radnice-Ovčín and Nižbor samples, and 307 Ma for Lobeč sample, respectively.

The sample Radnice-Ovčín came from a layer of a fine-grained kaolinite-rich volcanic tuff (the Bělka Horizon) of the Radnice Member (Fig. 3A). The sediment, which yielded a rich population of authigenic apatite grains, was taken from the wall of an abandoned coal mine situated to the south of Radnice (see Němejc 1953 and Opluštil *et al.* 2009 for the details on lithology and stratigraphy of this locality). Vitrinite reflectance measurements were made on coal particles taken from the Upper Radnice Coal Seam, which crops out approximately 10 m above the Bělka Horizon.

The Nižbor sample (Stradonice, Lísek) was collected from the Radnice Member. These deposits belonged to the lowermost part of the Carboniferous succession, which fills the Lísek erosional remnant, one of small, tectonically-constrained relict outliers of Carboniferous deposits that were preserved between Prague and Pilsen (Pešek & Martínek 2012). The sampling point was situated in a steep gorge on the right bank of the Berounka river, about 3 km to the SE of Nižbor (see Němejc 1953 and Holub & Obrhel 1967 for details). Apatite grains were extracted from the beds of coarse-grained conglomerates with arkose matrices. Thin coal seams and coaly claystone beds intercalating with the arkose provided vitrinite-rich materials sufficient for reflectance measurements.

The Lobeč sample was taken from coarse-grained arkoses stratigraphically belonging to the middle part of the Nýřany Member (Fig. 3B). The sampling point was located at the foothill of the Lobeč cliffs, exposed along the Vltava river at the northern periphery of Kralupy nad Vltavou (see also Opluštil *et al.* 2005 for geological details of this locality). While arkose sandstone itself provided a sufficient quantity of recycled detrital apatite grains, the partings of coaly claystone inter-bedded with arkose were rich in particles of dispersed organic matter suitable for reflectance studies.

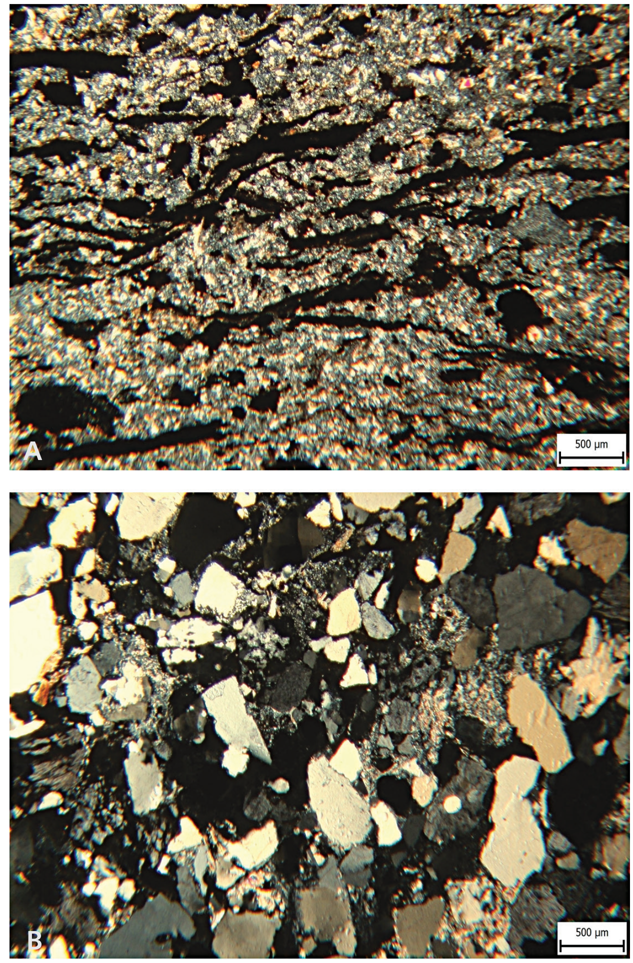

Figure 3. Photomicrographs showing aspects of representative Carboniferous apatite-bearing sediments (partlially crossed polars). • A – intensely kaolinized volcanic tuff (the Bělka Horizon) of the Radnice Member. The dark opaque seams and spots arranged parallel to bedding are coalified vegetal fragments. • B – coarse-grained arkose from the Nýřany Member, the Lobeč cliffs. Main rock components are angular and sub-rounded quartz and feldspar grains cemented by clay minerals. Some feldspar grains were partly replaced by sericite forming minute bright crystals.

Methods

Apatite fission track analysis (AFTA) and modelling

Apatite fission track (AFT) analysis is a technique for defining the thermal history of sedimentary rocks, including palaeo-temperatures and semi-quantitative estimates of palaeo-temperatures over time (Storzer & Selo 1984, Naeser *et al.* 1989). During the past 40 years, the method has gradually evolved into one of the most powerful tools to reconstruct the low-temperature thermal history of rocks, especially below ~120 °C, for tracks in apatite. The AFT analysis has been established as a particularly important instrument for basin analysis and hydrocarbon exploration because the apatite annealing temperatures between 60 and 120 °C essentially coincide with temperatures for liquid hydrocarbon generation (Armstrong 2005).

In uranium-rich apatite crystals, narrow damage trails (*i.e.* fission tracks) are formed by swift heavy ions ejected during spontaneous nuclear decay of ^{238}U. These tracks can be enlarged by chemical etching and counted using an optical microscope (Fig. 4). Track length distribution, in combination with apparent fission track ages, allows one to discriminate between different thermal histories, but proper interpretation of the data requires a complex mathematical treatment. Several software solutions have therefore been developed to compute thermal histories from AFT age and track length data (*e.g.* Laslett *et al.* 1987, Crowley *et al.* 1991; see also Armstrong 2005 for a review of the models). More recent overviews of fission track analysis techniques, including fundamentals of the method, have been provided by Gallagher *et al.* (1998), Gleadow *et al.* (2002), Tagami & O'Sullivan (2005), Lisker *et al.* (2009), Green & Duddy (2012) and Wagner & Van Den Haute (1992).

Apatite grain concentrates were obtained from ~7 kg rock samples using the standard heavy mineral liquid separation technique (Povondra & Ulrych 1988). The grains were mounted in Epofix®, polished and etched in 2.5% HNO_3 for 70 seconds to reveal the fission tracks. The external detector method (Wagner & Van Den Haute 1992) was applied for fission track analysis. A low-uranium muscovite detector (Jahre GmbH Wilhelmshaven, Germany) was fixed on a polished surface and stacked in an irradiation cassette with a CN5 glass neutron dosimeter. The cassette was irradiated with thermal neutrons in the TRIGA Mk. II Research Reactor at the Oregon University. A nominal fluence of thermal neutrons of $8.14 \times 10^{15} cm^{-2}$ was reached. Densities and lengths of spontaneous and induced tracks were measured by an Axioplan 3 (Zeiss) microscope coupled with an AUTOSCAN™ stage. The mean chloride content of apatite grains (*e.g.* Barbarand *et al.* 2003, Green & Duddy 2012) was determined using a CAMECA SX 100 Electron Micro Analyser. Fission track ages were calculated using the Zeta calibration factor according to Hurford & Green (1982, 1983). As age standards, apatite samples from Fish Canyon volcanic tuff, and Durango and Mount Dromedary intrusive complexes were used. The measured fission track densities were treated using the age-equation provided by Trackscan software (Tab. 1).

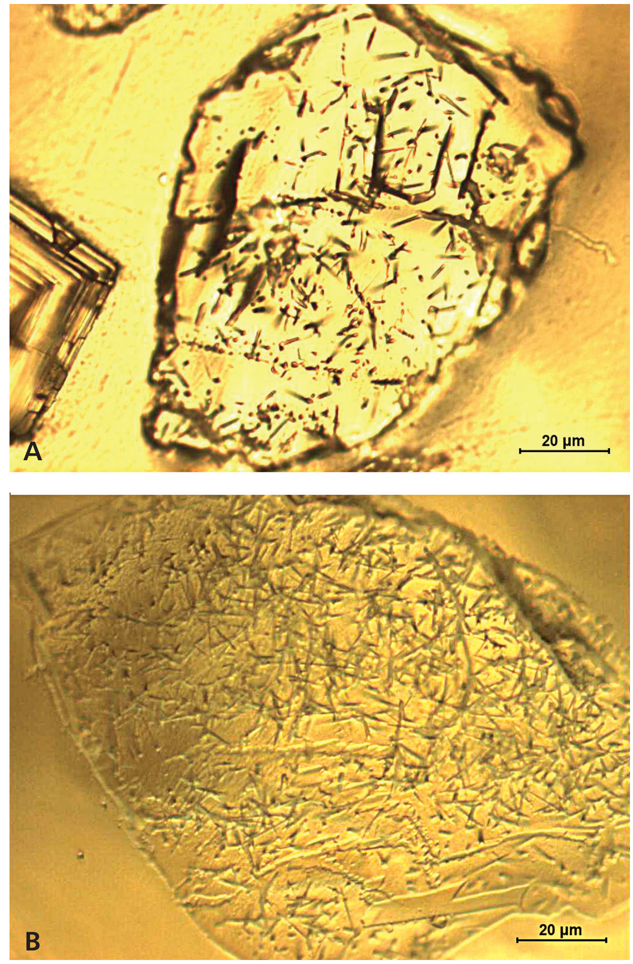

Figure 4. Microphotographs of fossil fission tracks in apatite grains separated from the Carboniferous sediments. Fission tracks were revealed by etching in 2.5% HNO_3. • A – angular authigenic apatite grain from the volcanic tuff (the Bělka Horizon) of the Radnice Member, Radnice-Ovčín quarry. The majority of linear features in the image are fission tracks. The difference in width of the tracks is due to a difference in etching rate that depends on crystallographic orientation. The latter is higher in the direction parallel to the c-axis. • B – subrounded detrital apatite grain from the arkose conglomerate, the Radnice Member, Nižbor. A number of fossil fission tracks can be recognized on the basal face of the crystal.

The thermal history of the rocks (*T-t* paths) was computed using the AFTSolve program (Donelick & Ketcham 1998; Ketcham *et al.* 2000, 2003, 2007; Ketcham & Apatite to Zircon, Inc. 2007), which uses the multi-kinetic annealing model (Carlson *et al.* 1999, Donelick *et al.* 1999, Ketcham *et al.* 1999). This model takes into account the variability in chemical kinetics between different apatite populations, including their chlorine content, which is known to exert a significant control on fission track annealing rates in apatite (*e.g.* Green & Duddy 2012 and the references therein). Chlorine contents are measured in every apatite grain, in which either age or fission-track length data are collected, and the AFTSolve model explicitly takes into account the influence of these Cl contents on annealing rates. This is one of the key advantages of the AFTSolve program that differs from other AFTA methods suggested by the end of the last century for interpretation of AFTA data.

Table 1. Analytical results of apatite fission track analysis for the samples from Central and Western Bohemian Carboniferous basins. Abbreviations: ρ_S – spontaneous track density of a sample; N_S – number of tracks counted to determine ρ_S; ρ_i – induced track density measured in a muscovite external detector; N_i – number of tracks counted to determine ρ_i; n – number of counted grains; $P(\chi^2)$ – Poisson distribution test at 95% confidence level; TL – mean length of horizontal confined fission tracks with standard deviation in all crystallographic orientations; N – number of measured confined tracks; σ – standard deviation for the length distribution of confined fission tracks; Cl – chlorine content in studied apatite grains.

Sample code	Locality	Lithology	AFT-age [Ma + 1σ]	ρ_s (N_s) [$\times10^6$ cm^{-2}]	ρ_i (N_i) [$\times10^6$ cm^{-2}]	n	P(χ^2) [95%]	TL (N) [μm]	σ [μm]	Cl
K3	Lobeč	arkose	204 ± 25	4.77 (1360)	3.19 (939)	6	fail	11.6 (90)	1.8	< 0.1
L2	Nižbor	arkose	172 ± 17	5.85 (5482)	4.42 (4219)	20	fail	12.5 (154)	1.5	< 0.1
H11	Radnice-Ovčín	volcanic tuff	264 ± 7	6.89 (6332)	7.10 (6528)	20	fail	12.2 (94)	1.8	< 0.1

The Monte Carlo method was applied to test 10,000 to 20,000 possible *T-t* paths (Ketcham *et al.* 2000). During the modelling process, the temperature and time constraints were kept at the minimum. Only the minimum temperature above the total annealing temperature of the most resistant apatite species, the sample age and the surface temperature of the sample from which apatite was collected were introduced as the earliest and final *T-t* constraints, respectively (*e.g.* Ketcham *et al.* 2000). Further details on the modelling process can be found elsewhere (Filip & Suchý 2004).

Vitrinite reflectance (VR) measurement, modelling, and its integration with the AFTA data

The technique of vitrinite reflectance measurement is now commonly used to determine the thermal maturity of coal and hydrocarbon source rocks ranging from low grade diagenesis to very low grade metamorphism, *i.e.* from ~40 °C to ~400 °C (Hunt 1979, Tissot & Welte 1984; see Teichmüller 1987, Suarez-Ruiz *et al.* 2012, Hartkopf-Fröder *et al.* 2015, Ferreiro Mählmann & Le Bayon 2016 for reviews). The increase in VR is dependent on many parameters, including temperature, geologic time, fluid chemistry, pore fluid pressure, oil content and degree of tectonic deformation (*e.g.* Hood *et al.* 1975; Barker & Pawlewicz 1994; Huang 1996; Dalla Torre *et al.* 1997; Suchy *et al.* 1997; Le Bayon *et al.* 2011, 2012), but the influence of maximum temperature and duration of heating are the most important (Teichmüller 1987, Mukhopadhyay 1992). The technique of VR measurement has significant advantages over other geo-thermometers, such as clay minerals, fluid inclusion, and fission track annealing in that the vitrinite is commonly present in many sedimentary rocks, its reflectance is irreversible and rapidly stabilizes in response to the maximum temperature to which the entire system has been exposed (Price 1983, Barker & Pawlewicz 1994; see also Tobin & Claxton 2000 and the references therein). Compared with mineral indices, vitrinite reacts faster to heating (Ferreiro Mählmann *et al.* 2012, Potel *et al.* 2016).

Using reaction kinetics (Arrhenius equations), the relationship between VR, rock temperature, and heating time can be used to reconstruct geothermal history (Buntebarth & Stegena 1986, Robert 1988). Early kinetic models of vitrinite maturation applied to petroleum exploration used a simple concept in which the rate of maturation doubles every 10 °C (Lopatin 1971). Since then, a number of more sophisticated kinetic models of vitrinite maturation have been proposed, as reviewed by Waples (1980, 1989), Wood (1988), Morrow & Issler (1993), Le Bayon *et al.* (2012), Burnham *et al.* (2017) and Nielsen *et al.* (2017).

The kinetics of vitrinite reflectance (VR) is generally similar to that of fission track annealing in apatite (Burnham & Sweeney 1989). This makes VR an ideal complement to the AFTA technique when applied to sedimentary sequences (see Green & Duddy 2012 and the references on combined AFTA/VR studies therein). The combination of AFTA and VR data has two distinct advantages (Bray *et al.* 1992, Arne & Zentilli 1994, Duddy *et al.* 1998): firstly, the two techniques, which are mutually independent, can be considered as maximum recording thermometers. This provides a valuable check on maximum palaeo-temperatures. AFTA data give palaeo-temperature estimates of up to a maximum value, usually around 120 °C, at which total annealing occurs in apatite, whereas VR continues to react to substantially higher temperatures. Thus, VR combined with AFTA data can be of great assistance in confirming or revealing thermal events, which may not be confidently defined by AFTA alone (*e.g.* Green & Duddy 2012, Green *et al.* 2002). Secondly, the VR variations themselves bear no time information at all, but using the Sweeney & Burnham (1990) model, an agreement can be achieved between observed and predicted VR values through the thermal history time, using the framework provided by AFTA data. This is particularly important, because data from either technique alone might be viewed with discretion, but when two independent techniques provide comparable palaeo-temperatures, the conclusions can be regarded as reliable.

Reflectance data were collected on macroscopic coaly particles from thin coal seams (Radnice-Ovčín and Stradonice) and vitrinite fragments dispersed in sedimentary rocks (Lobeč). The samples were cut and polished normal to bedding and studied using an Olympus BX51 optical microscope equipped with a Zeiss Photomultiplier MK3 under both reflected-light and fluorescence regimes using lenses with ×40 and ×100 magnifications. The Pelcon point counter was used for maceral analysis. The determinations of vitrinite, liptinite and inertinite group macerals were executed following the recent ICCP recommendations (*i.e.* ICCP 1998, 2001; ISO 7404-3 2009; Taylor *et al*. 1998; Pickel *et al*. 2017). Random reflectance values of organic matter (R_r %) were determined using the Spectra Vision software. Sapphire (R = 0.596%), yttrium-aluminium-garnet (R = 0.894%), and N-LASF46A (R = 1.309%) reflectance standards were used for calibration of measurements according to ISO 7404-5 (2009) and Taylor *et al*. (1998). The number of reflectance measurement points (n) varied from 20 to 100 measurements per sample ($n_{average}$ = 82), with standard deviations (σ) ranging from 0.04 to 0.09; Tab. 2).

The VR modelling was applied to resolve three mutually interrelated tasks: (1) to reconstruct early stages of thermal history of the samples including maximum palaeo-temperatures that cannot be estimated by the AFTA method alone, (2) to provide an independent check of AFTA-derived *T-t* paths of the samples, and (3) to estimate the timing and duration of coalification. The VR modelling was used to reconstruct early stages of the thermal evolution of L2 – Nižbor and K3 – Lobeč samples, which occurred before the annealing of its apatite fission tracks. For H11 – Radnice-Ovčín sample, which was exposed to temperatures <120 °C, the VR modelling was employed merely as an independent check of its *T-t* path derived from AFT data.

The time-temperature evolution of vitrinite was simulated by repeated iterative runs of the EASY%R_o program (Sweeney & Burnham 1990). This vitrinite maturation model applies the kinetic variables, activation energy, and frequency factors estimated using different heating rates during Rock-Eval pyrolysis (Fig. 5). It can be conveniently implemented on a spreadsheet, or in the form of a small program on a personal computer. The model can be used with any type of thermal history including non-deposition, uplift and cooling, and for VR values ranging from 0.3 to 4.5% R_r, and for heating rates ranging from those in the laboratory (1 °C/week) to those in slowly subsiding basins (1 °C/10 m.y.). For each sample studied, multiple combinations of temperatures and heating times were tested until a reasonable fit with the measured R_r values was reached. The original input data (*i.e.* temperatures, heating times) and resulting VR values obtained during individual EASY%R_o runs are available on request from the first author as Excel spreadsheets.

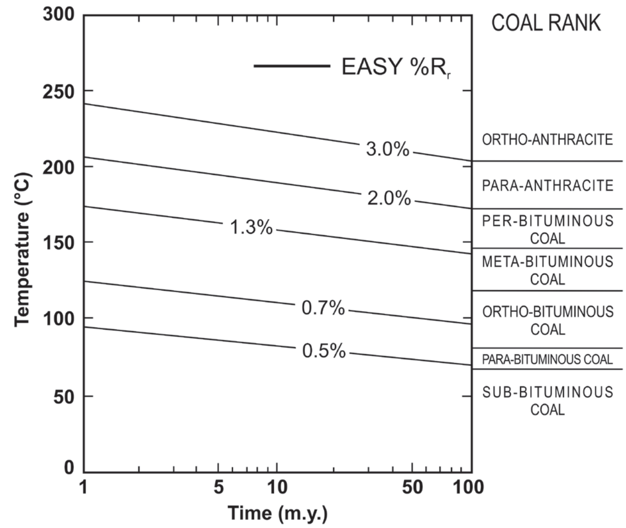

Figure 5. Nomogram of vitrinite reflectance *vs*. exposure time and maximum temperature calculated using EASY%R_o (after Sweeney & Burnham 1990, with modifications). Correlation between vitrinite reflectance and coal rank of seam coals (after International Classification of in-Seam Coals 1998) is also shown.

We deliberately limited our study to a simple reconstruction of thermal evolution of the samples, without making any attempt to retrieve its burial and/or subsidence histories. This minimalistic approach was primarily caused by a very limited amount of reliable information that we were able to collect on the thicknesses of stratigraphic overburden at the localities under study, as well as on organic matter reflectance values from overlying Cretaceous sediments. There is a hope that in the future, when more background data are available, full-scale comprehensive regional study will be possible, including a detailed burial history reconstruction of Carboniferous sediments.

Results and Interpretations

Apatite fission track analysis

Apatite fission track analysis (AFTA) of all three samples revealed that its confined tracks of apatite grains were shortened from their initial length of 16.3 μm to about 11.6 μm (Tab. 1). This implies that the samples experienced a partial (H11 – Radnice-Ovčín) to complete annealing (L2 – Nižbor and K3 – Lobeč) during their palaeo-thermal histories. In order to fully elucidate the meaning

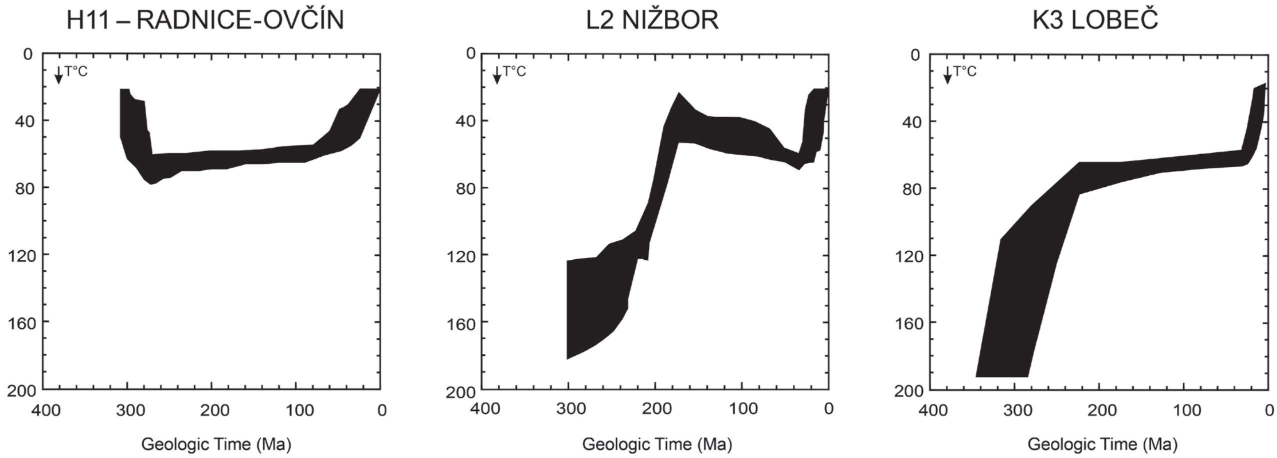

Figure 6. Results of reverse thermal modelling of the apatite fission tracks for three samples from Central and Western Bohemian Carboniferous basins using the AFTSolve software. The black areas encompass all good and best-fit *T-t* paths with a probability of more than 90%. Note that temperatures in excess of 120 °C represent a software artefact, with no geological meaning (see Ketcham *et al.* 2000 for AFTSolve programme details.).

of the data, thermal histories of the samples were modelled based on available thermo-chronological constraints (Fig. 6).

The H11 – Radnice-Ovčín sample of volcanic tuff never entered into the zone of complete annealing. As a consequence, the sample preserves a complete *T-t* record from the moment of apatite grain deposition until recent time. It shows that the sediment was intensely heated soon after deposition and exposed to maximum palaeotemperatures of about 75 °C that culminated during the early Permian time. Starting from ~280 Ma till about 60 Ma, the rock experienced a long period of thermal stagnation, during which, it resided in a zone of stable temperatures of about 60–70 °C. This part of the thermal history of the sample correlates well with the known regional sedimentary record, which evidences low sedimentation rates and/or non-deposition over a substantial part of the Mesozoic (*e.g.* Malkovský 1979). Rapid cooling of the sediments began about 40 Ma. This thermal event can be paralleled with a major structural inversion that has affected the interior of the Bohemian Massif since the Paleogene period (Malkovský 1979, 1987; see also Ziegler *et al*. 1999, Zeman *et al.* 2000, Filip & Suchý 2004).

The modelling of *T-t* paths for Nižbor (L2) and Lobeč (K3) samples showed that these sediments were subjected to temperatures in excess of ~120 °C, which resulted in the total annealing of apatite grains before ~200–260 Ma (Permian to Early Jurassic).

Unfortunately, the AFTA method alone does not allow for the reconstruction of thermal evolution preceding annealing. The post-annealing thermal history of the samples, however, can be reconstructed and it shows that the rocks experienced two-stage cooling (Fig. 6). The first cooling episode occurred when the sediments ascended from the zone of total annealing. This temperature drop was probably due to post-Variscan structural inversion and erosion that removed a part of Carboniferous–Permian sequence in the area (*e.g*. Holub *et al.* 1997), but a steep decrease in regional heat flow during the Permian to Jurassic period may also have been involved (see also vitrinite reflectance modelling below). This first cooling episode terminated at 180–220 Ma (Jurassic–Triassic). Since then, until about 20–40 Ma, the thermal histories of both samples were essentially similar to those of the Radnice-Ovčín (H11) sample, including a period of prolonged Mesozoic to Cenozoic thermal stagnation at 60–70 °C, followed by fast cooling to surface temperature during the Paleogene–Neogene time. A minor temperature increase that can be seen in the *T-t* path of L2 Nižbor sample for a period around 90–40 Ma can be probably attributed to heating beneath several hundred metres of overburden of Cretaceous to (?)Paleogene sediments that may subsequently have been removed. Some previous studies have already indicated that the Cretaceous sedimentation in the area may have continued locally after the Santonian, and a thickness of more than to 1,000 metres of sediments may have been eroded from the top of the existing basin fill (*e.g*. Uličný & Franců 1996, Holub *et al*. 1997).

Vitrinite reflectance measurement and modelling

Vitrinite reflectance values ascertained in the Carboniferous sediments are presented in Tab. 2. The values of random reflectance (R_r) of vitrinite ranged from 0.53 in the SW (Radnice-Ovčín) to 0.83% R_r in the NE (Lobeč). These reflectance values are generally indicative of para-bituminous to ortho-bituminous medium-rank coals (International Classification of in-Seam Coals 1998).

The coal samples taken from the Upper Radnice Coal Seam at the Radnice-Ovčín quarry represent dull-banded coal with a low content of mineral admixtures (Fig. 7A). The coal consists of common vitrinite-group macerals, with subordinate liptinite and inertinite particles. The dominant vitrinite maceral present is colotelinite on which the reflectance measurements were performed.

Thin coal seams and dark claystone partings sampled at Nižbor yielded a similar assemblage of organic constituents dominated by vitrinite-group macerals. Colotelinite and less abundant colodetrinite were identified as the most common types of vitrinite group materials in these samples, along with subordinate particles of liptinite and inertinite groups (Fig. 7B).

Dark coaly claystone samples from Lobeč cliffs were found to be relatively poor in recognizable organic particles. Scarce detrital vitrinite and inertinite fragments were found dispersed in a clayey matrix or embedded in secondary quartz cement (Fig. 7C, D). Additional details on VR measurements and organic-petrological characteristics of individual samples are summarized in Tab. 2.

Figure 8 shows the calculated VR values and the respective palaeo-temperatures of individual samples as they evolved from the deposition time to the present. At Radnice-Ovčín, the modelling of an average VR value

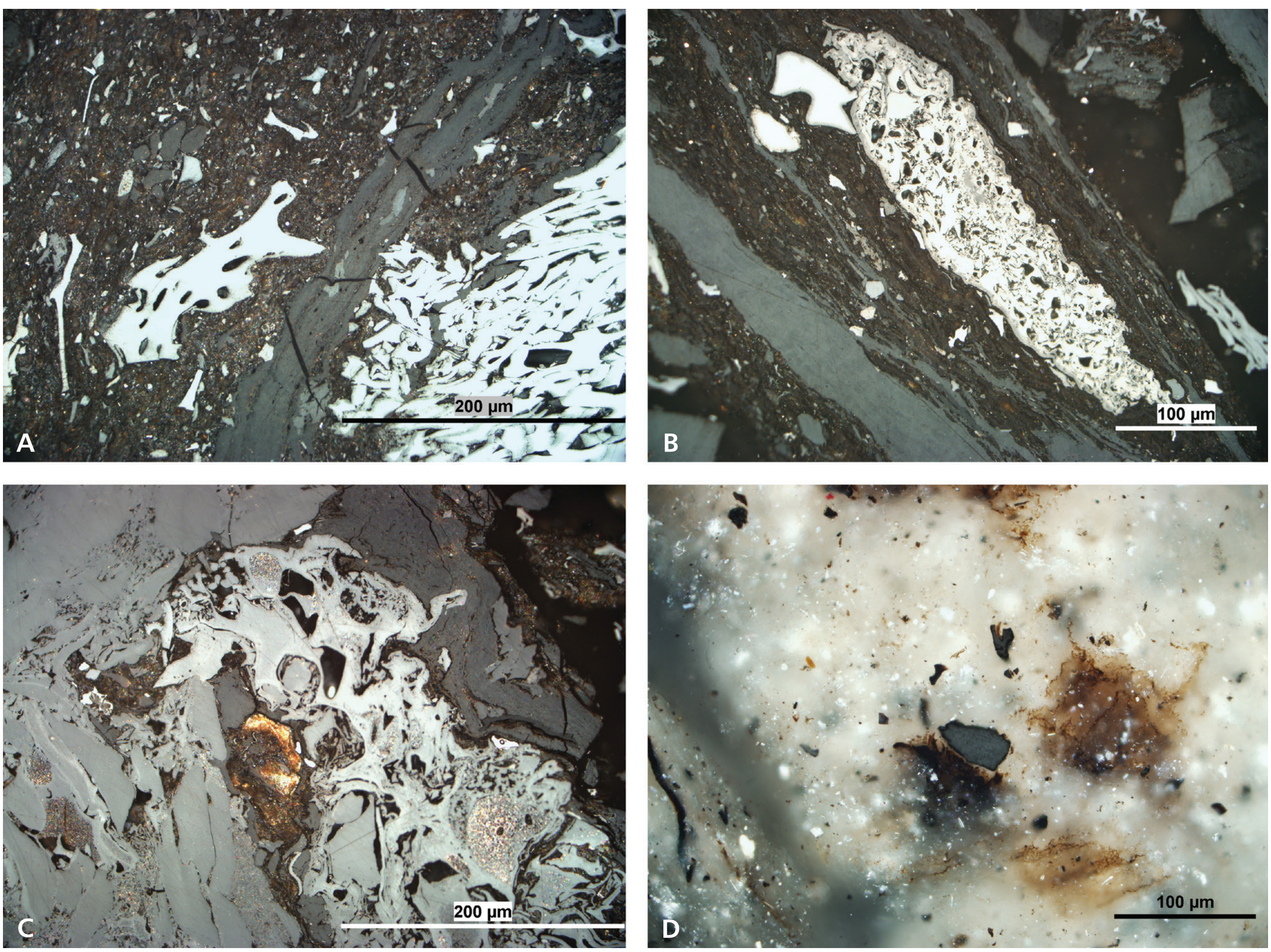

Figure 7. Microphotographs showing characteristic types of organic macerals recognised in Carboniferous sediments. Microphotographs were taken on polished rock surfaces under oil immersion, and normal reflected light. • A – elongated band of medium-grey vitrinite (colotellinite) surrounded by bright whitish grains of inertinite materials (fusinite and semifusinite). Detrital matrix is rich in small liptinite fragments, finely disseminated clay minerals and vitrinite and inertinite fragments. The Upper Radnice Coal Seam, Radnice-Ovčín quarry. • B – elongated medium grey bands of vitrinite (colotellinite) in association with brightly white reflecting intertinite-group particles (semifusinite, macrinite and inertodetrinite) bound in a fine-grained detrital matrix rich in liptinite fragments. Nižbor. • C – medium-grey vitrinite fragments (left) and a larger grey inertinite particle (centre). Brown-coloured small grains are liptinites. Lobeč. • D – small medium-grey vitrinite particles embedded in massive whitish quartz cement. Diffusely outlined, rusty brown bodies are clay minerals. Lobeč.

Table 2. Organic-petrological data of Carboniferous coal-bearing sediments. See text for details.

Sample No.	Locality	R_r (%)	σ	R_{rmin}–R_{rmax} (%)	Number of measured points (n)	Vitrinite (vol. %)	Liptinite (vol. %)	Inertinite (vol. %)	Minerals (vol. %)
98/089	Radnice-Ovčín	0.53	0.04	0.48–0.60	100	62.1	15.8	12.6	9.5
03/249	Radnice-Ovčín	0.58	0.09	0.48–0.67	100	50.2	30.8	11.4	7.6
98/085	Radnice-Ovčín	0.65	0.08	0.56–0.72	100	46.1	22.0	18.8	13.1
	Radnice-Ovčín (average value per locality)	*0.59*							
43/211	Nižbor	0.73	0.08	0.65–0.80	60	50.6	8.2	10.5	30.7
98/086A	Nižbor	0.63	0.08	0.58–0.74	100	42.2	24.6	15.1	18.1
98/086B	Nižbor	0.72	0.06	0.65–0.78	100	67.5	8.3	9.4	14.8
	Nižbor (average value per locality)	*0.69*							
14/001	Lobeč	0.77	0.09	0.65–0.85	55	9.3	7.4	13.8	69.5
14/001B	Lobeč	0.83	0.09	0.74–0.92	100	10.4	8.7	15.1	65.8
14/002	Lobeč	0.73	0.08	0.63–0.81	20	1.2	4.1	1.3	93.4
	Lobeč (Kralupy) (average value per locality)	*0.77*							

(R_r = 0.59%) resulted in a vitrinite *T-t* path, that is very close to that provided by the AFTA data. According to the EASY%R_o algorithm, the sediment experienced a substantial and relatively fast heating soon after its deposition. The coalification corresponding to the level of sub-bituminous coals (R_r ~ 0.40%) was largely accomplished during the Carboniferous–early Permian time, from 15–40 m.y. after the deposition of the peat. It should be noted, however, that the EASY%R_o model also allows for an even faster coalification progress in which the VR values of 0.40% R_r would have been completed as soon as 4 m.y. after the peat deposition. The maximum palaeo-temperatures of around 85–90 °C, and the respective VR values between 0.49–0.56% R_r, were attained 35–40 m.y. after deposition (*i.e.* during the early Permian time). The modelling also indicates that the

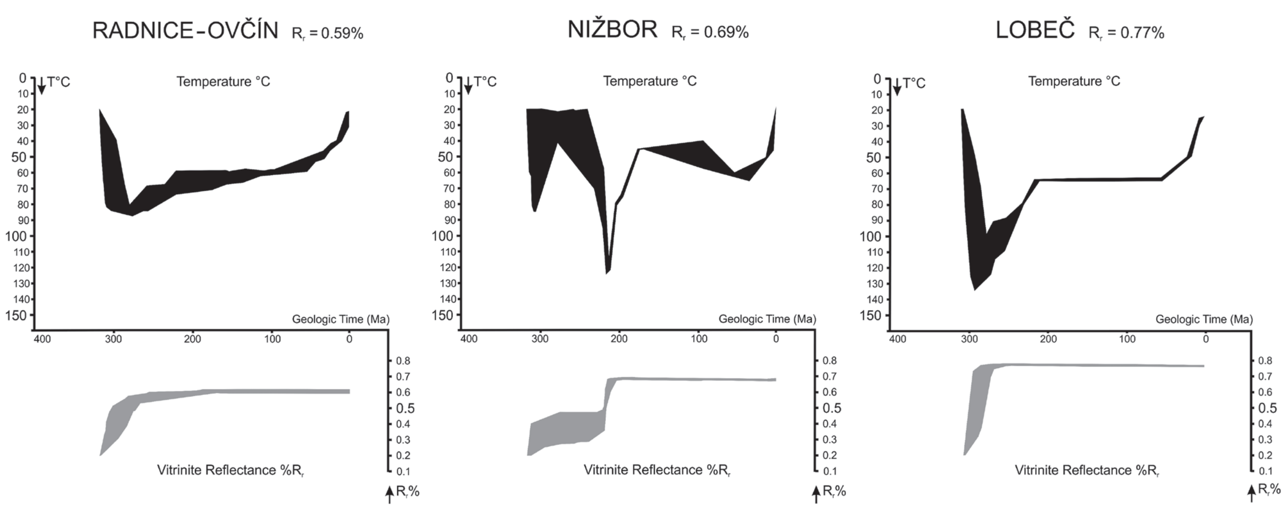

Figure 8. Thermal modelling of vitrinite reflectance using the model of Sweeney & Burnham (1990). Variable input time-temperature history data are shown as black envelopes. Corresponding vitrinite reflectance values modelled with EASY%R_o software are shown in grey. Note a shift in maximum palaeo-temperatures between the samples that gradually increase from SW (Radnice-Ovčín) to NE (Lobeč). See the text for details.

subsequent Mesozoic episode, when the sample resided for nearly 200 m.y. in the zone of thermal stagnation at 60–75 °C, had only a limited effect on the final VR value.

The modelling of elevated VR values that were ascertained at two other eastward localities (*i.e.* 0.69% R_r at Nižbor and 0.77% R_r at Lobeč), suggests maximum palaeo-temperatures of 125 °C and 135 °C, respectively. This is in a general agreement with the results of AFTA modelling that indicate a complete annealing of apatite fission tracks of these two samples due to palaeo-temperatures in excess of ~120 °C before 200 Ma and ~260 Ma, respectively.

The modelling of the pre-annealing thermal evolution of the Nižbor sample allows for two different end-member *T-t* scenarios. Firstly, the sediment was rapidly and intensely heated to ~80 °C from 4 to 10 m.y. after its deposition, in a way similar to the Radnice-Ovčín sample. During this early thermal event, which may have occurred during sedimentation of the Nýřany Member, the VR values were raised to around 0.40% R_r, corresponding to sub-bituminous coal. This was followed by cooling to about 40 °C, which was achieved about 40 m.y. after deposition (*i.e.* during the early Permian). Then, upon the subsequent second-stage heating that culminated around the Triassic-Jurassic transition, the maximum temperature of about 125 °C was achieved. An alternative end-member scenario also exists that the sample experienced only a simple one-stage gradual heating during the Carboniferous to Triassic time, which culminated in maximum temperatures being attained at about 220 Ma. The vitrinite modelling also indicates that additional moderate heating, which occurred during the Cretaceous to Paleogene times (*i.e.* between ~100–40 Ma), had very little, if any, impact on resulting VR values; the vitrinite had already been largely stabilized before ~200 Ma.

For the Lobeč sample, the vitrinite modelling predicts steeply increasing palaeotemperatures during the first 20 m.y. after sediment deposition. This resulted in a significant VR increase so that the R_r values of around 0.40% were achieved already during the Carboniferous Period. The EASY%R_o software predicts that these elevated VR values could have been attained extremely soon, about 2–4 m.y. after sediment deposition. The maximum palaeo-temperatures of ~135 °C that largely induced the present-day VR values culminated during the early Permian (early Cisuralian) time, 270–290 Ma.

Discussion

The vitrinite reflectance modelling showed that the Carboniferous sediments were exposed to palaeo-temperatures, which ranged from ~85 °C (Radnice-Ovčín) to ~135°C (Lobeč). Although the kinetic modelling of vitrinite reflectance predicts an array of possible temporal scenarios, much of our data strongly indicate that maximum heating occurred soon after the deposition of Carboniferous strata, during the Carboniferous or early Permian times. Both apatite fission track and vitrinite reflectance modelling confirm a significant drop in sediment temperature before the onset of the Mesozoic thermal stagnation period and, a second prominent period of fast cooling that occurred during the Cenozoic Era. Remarkably similar palaeo-geothermal evolution has already been documented for Lower Palaeozoic and Neoproterozoic rocks of the Barrandian area, immediately to the south of the Carboniferous basins (Filip & Suchý 2004; Suchý *et al*. 2007, 2015). This implies that the Carboniferous basins and their underlying geological units experienced a similar thermal evolution that was apparently controlled by regional geological processes.

Our data, though limited in extent, indicate that across the area, the coalification grade of Carboniferous coals slightly increased from SW to NE. Similar regional trends in coalification grade and palaeo-geothermal gradients have been already reported for Central–Western Bohemian Carboniferous deposits in some previous studies (*e.g.* Šafanda *et al.* 1990, Pešek 1996,), but the reason for these variations remain speculative. In general, palaeo-temperatures recorded in Carboniferous sediments and regional changes across the area can be explained either by variations in the original thickness of stratigraphic overburden, or by variations in paleo-heat flow, or by a combination of both.

The present-day Carboniferous overburden does not exceed several tens of metres. The original thickness of Carboniferous and/or Permian overburden remains unknown since all three localities are situated along erosive margins of the basins where the removal of overlying sediments was probably intensive. The average thickness of preserved Carboniferous–Permian overburden extrapolated from the interior parts of the basins (*i.e.* about 700–900 m), even if combined with several hundreds of metres of hypothetical Cretaceous sedimentary cover (*e.g.* Holub *et al.* 1997, Martínek *et al.* 2017) is clearly insufficient to explain the thermal maturity of the Carboniferous sediments, at least under the present-day geothermal gradient (~30 °C/km up to 35 °C/km; Šafanda *et al*. 1990).

Indeed, only heat flows that were elevated in the past can provide a plausible mechanism to explain the maximum palaeo-temperatures as ascertained in the sediments. Dvořák (1989), Dvořák & Paproth (1988) and Zwart (1969, 1975) have suggested palaeo-gradients as high as 200 °C/km for some parts of the Variscan orogen in Central–Western Europe. Botor & Anczkiewicz (2015), Littke *et al.* (1994) and Robert (1989) have also documented elevated Variscan heat flows for some coal-bearing basins of Western Europe (see also Littke *et al*. 2000 and their discussion of the Permo-Carboniferous heat flow). Furthermore, the late Carboniferous to early Permian thermal climax has already been proposed to affect the diagenesis of Ordovician to Devonian sediments in the Barrandian area, immediately to the south of the Carboniferous basins (Franců *et al*. 1998, Glasmacher *et al*. 2002).

In this study, elevated Carboniferous–Permian heat flows are newly corroborated by vitrinite reflectance modelling, which suggests a steep palaeo-temperature rise soon after sediment deposition. The precise calculation of Carboniferous thermal gradients is, however, problematic as a substantial part of overlying Carboniferous strata and much of younger sedimentary cover were removed by erosion and its original thickness remains a matter of debate. Numerous stratigraphic breaks within the Carboniferous sequence, some of which lasted between 1–3.6 m.y. (*e.g*. Opluštil *et al.* 2016) provide additional uncertainties related to the original thickness of Carboniferous sediments that may have been deposited and/or eroded during these stratigraphic breaks. Nevertheless, simple conservative estimates show that the thermal gradients during the Carboniferous must have indeed been very high, probably around 200 °C/km, or even higher, similar to those of some present-day active rifts or geothermal fields (*e.g*. Palmer *et al.* 1975, Dornstadter *et al.* 1999).

The high Variscan heat flows could have been due to a number of factors including the thinning of the underlying lithosphere, abundant volcanic activity (*e.g.* Dvořák & Paproth 1988) or hot igneous intrusions that penetrated into the subsurface. Klomínský (1994) has pointed out that the Variscan heat flow in the Bohemian Massif may have been substantially elevated by a number of granitic plutons of variable size, many of which were emplaced around 350–307 Ma (see also Henk *et al*. 2000 for their discussion on the role of granitoid intrusions in high-temperature metamorphism in the Variscan Orogen of Central Europe). However, granitic bodies are not present close to any of the localities examined. Gravity Bouguer anomalies around Kralupy nad Vltavou, close to the Lobeč locality (Sine 2000), may indicate the presence of plutonic bodies buried shallowly below the surface, but the age of these hypothetical intrusions, if any, remains unknown. Rhyolite and basaltic igneous intrusions of variable thicknesses are known to occur in the sediments of the Radnice Member and the Líně Formation (Pešek 2004). Abundant volcanic effusives intercalating with Carboniferous sediments also point to widespread syn-sedimentary volcanic activity. Unfortunately, volcanic centres, that produced these effusive deposits, are either fully covered by younger sediments or have been eroded in the past (*e.g*. Mašek 1973) so that their contribution to the ancient heat flow balance is difficult to quantify.

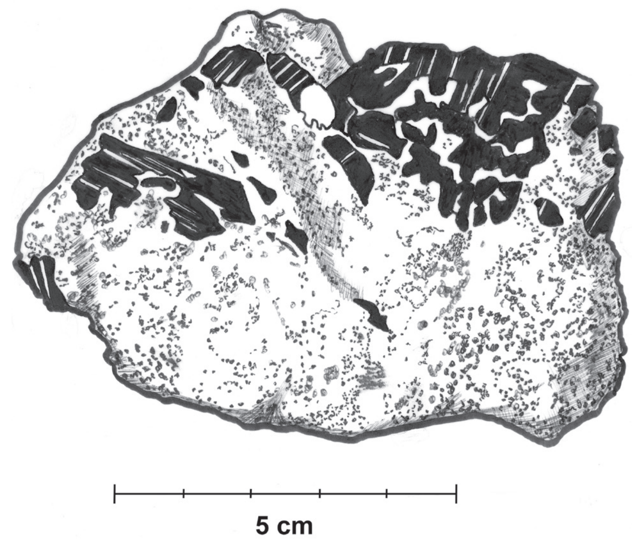

Figure 9. Drawing of a sample of redeposited coal clasts embedded in kaolinite-rich sandstone matrix (modified after black and white photograph from Pešek & Sýkorová 2006). Note numerous cleats filled with clay minerals that cut through coaly fragments, perpendicular to its original bedding. These structural relationships evidence a very early onset and short duration of the coalification process; Kladno-Rakovník Basin.

Except igneous intrusions or fossil volcanic activity, heat transport due to hot fluids migrating through fractures or along permeable horizons may have also been responsible, at least in part, for high palaeo-temperatures recorded in the Carboniferous sediments. These hot fluids may have contributed to the rapid coalification of organic matter enclosed in sedimentary strata (*e.g*. Daniels *et al*. 1990; Gayer *et al*. 1996, 1998; Marino *et al*. 2015; see also Duddy *et al.* 1998 and Hower & Gayer 2002 for the reviews.). Whether the Carboniferous sediments have ever been exposed to heated fluids remains uncertain, but some geological observations may point to such a possibility. In particular, basal Carboniferous sediments in the Kladno-Rakovník basin were locally penetrated by quartz-carbonate veinlets that hosted aqueous inclusions of variable salinity. These palaeo-fluids were entrapped at temperatures of 156–187 °C, which were substantially higher than maximum burial temperatures inferred for the Carboniferous strata from other palaeo-thermometers (Zachariáš & Pešek 2010, 2011). In a similar way, Suchý *et al.* (2017) have recently documented coaly particles yielding vitrinite reflectance values substantially elevated above the burial diagenetic backgrounds that were concentrated along goethite, gypsum, and jarosite-mineralized bedding planes and tectonic fractures in the Nýřany Member strata. They interpret this relationship as evidence that the Carboniferous sediments were exposed to hot mineralized fluids flowing through fractures or along permeable bedding planes. Similar examples of the diagenetic grade being escalated by fracture-bound migration of heated basinal fluids have already been recognized in Silurian and Devonian sediments of the nearby Barrandian area (Volk *et al*. 2002, Suchý *et al*. 2010).

Whatever the mechanisms driving elevated palaeotemperatures in the Carboniferous sequence, it follows that these strata must have been heated relatively soon after deposition when the sediments were still buried shallowly below the surface. This was clearly evidenced by numerous occurrences of redeposited clasts of sub-bituminous coals that occur at various stratigraphic levels of the Carboniferous sequence in the Plzeň and Kladno-Rakovník basins (Pešek 1978, Daněk *et al*. 2002). Many of these coaly clasts reveal abundant cleats oriented perpendicular to the original coal bedding. This strongly indicates that the coalification must have occurred prior to coal redeposition (Fig. 9). By combining structural observations with wider stratigraphic knowledge, some researchers have already concluded that the process of coalification occurred at a shallow burial level (Pešek & Sýkorová 2006). These studies indicate that despite a thin sedimentary overburden, the peat-to-coal transformation proceeded extremely rapidly, with the coalification duration ranging from tens or several hundred thousands of years to a maximum of 2 m.y. Abundant stratigraphic hiatuses that were newly recognized within the Carboniferous succession (*e.g*. Opluštil *et al*. 2016) also indicate that the Carboniferous strata must have been periodically eroded, thus allowing for exhumation and erosion of some shallowly buried coal seams.

Our vitrinite reflectance modelling provides new independent evidence that VR values around 0.4% R_r, indicative of a sub-bituminous coal rank, were attained very rapidly, as soon as 2 to 4 m.y. after peat deposition. An even more rapid increase and stabilization of vitrinite reflectance has been confirmed in other studies of natural open, fluid-rich geothermal systems and explained in terms of the effects of rapid thermal pulses that existed for only 10^0–10^4 years (Barker 1991, Barker & Pawlewicz 1994, Le Bayon *et al.* 2011). It should be noted, however, that the Burnham & Sweeney (1989) model that we used for vitrinite modelling was restricted in that it did not allow for simulation of thermal events shorter than 1 m.y.

Conclusions

Our research using apatite fission track analysis and vitrinite reflectance kinetic modelling as complementary thermo-chronometers provides the following insights into the palaeo-geothermal history of Upper Carboniferous sediments of Central and Western Bohemia:

(1) During the Variscan Permo-Carboniferous thermal climax, geothermal gradients were extremely elevated (~200 °C/km or even more), so that coalification proceeded rapidly and a substantial coal rank was reached close to the surface. Under these harsh geothermal conditions, coalification of the peat was completed within ~2–4 m.y. after deposition, or even earlier. During depositional hiatuses some shallowly buried coal seams were eroded and their remains were redeposited as intraclasts into younger Carboniferous sediments.

(2) The AFTA data shows that the sediments were exposed to maximum temperatures ranging from ~75 °C in the SW to more than 120 °C in the NE, and these were attained prior to ~200–260 Ma (Permian to Early Jurassic time). This initial heating stage was followed by a period of regional cooling and thermal stability at temperatures between ~50–75 °C, lasting throughout much of the Mesozoic and Cenozoic eras. A second episode of accelerated cooling and erosion began at ~20–40 Ma and caused the present exposure of rocks at the surface.

(3) Vitrinite reflectance modelling using average VR values of 0.59% R_r in the SW and 0.77% R_r in the NE, corroborates the AFTA data and provides additional information on the pre-Jurassic evolution of the sediments. The modelling predicts heating at maximum temperatures ranging between ~85–135 °C that rapidly ascended during several millions of years after deposition of the sediments. This early heating caused a steep increase in vitrinite reflectance to values of ~0.40–0.50% R_r, which were largely achieved during the Carboniferous time.

(4) The cause for high heat flows that dominated during the Carboniferous–Permian periods was probably intense syn-sedimentary volcanism and/or igneous activity combined with heat transport provided by hot fluids that circulated through the strata.

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