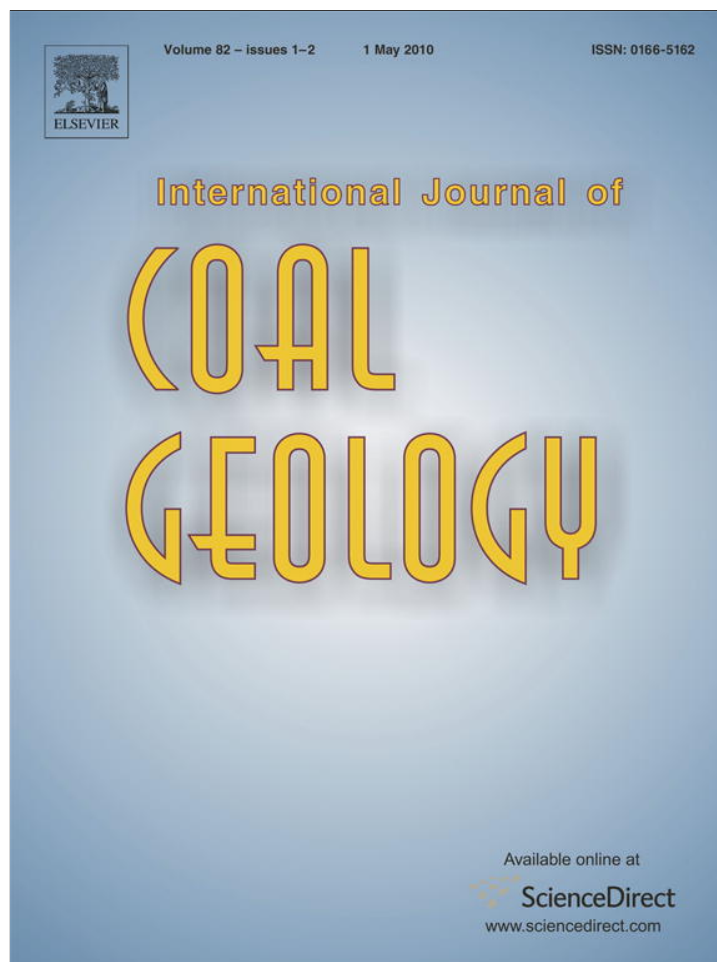


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Calculation of the conodont Color Alteration Index (CAI) for complex thermal histories

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ABSTRACT

A simple model for the Arrhenius reaction conodont Color Alteration Index (CAI) that can be implemented easily in a computer program is introduced. The model, named to as EasyCAI, envisages the overall process of conodont alteration as a series of 12 parallel pseudo-first-order reactions. Our approach is especially amenable to a spreadsheet program and can solve complex geological histories involving variable heating and cooling rates. With EasyCAI, a profile of CAI values versus time can be obtained for a given stratigraphic level if the time–temperature history for that level has been estimated. EasyCAI can provide additional constraints to optimize thermal history models by computing profiles of CAI values with depth and time; thus providing a new tool for quantitative CAI palaeothermometric analysis.

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1. Introduction

Conodonts are tiny (0.2 to 2 mm) apatitic remains of the feeding apparatus of an extinct animal group of the Class Conodonta, which was common in Cambrian to late Triassic oceans. These tooth-like microfossils have become a widely used tool in integrated basin analysis and hydrocarbon exploration because of its coupled use as precise biostratigraphic markers and thermal maturity indicators. The latter property is based on the analysis of color that changes as the conodont elements experience progressive and irreversible chemical transformations of the organic matter interspersed within the hyaline crown tissues (Marshall et al., 2001; Trotter et al., 2007) as a response to increasing temperature with time. The conodont color change has long been known (Ellison, 1944) but it has not been explained and quantified until the works of Epstein et al. (1977) and Rejebian et al. (1987), who established the conodont Color Alteration Index (CAI) and calibrated their values according to heat and exposure time, in accordance with the Arrhenius reactions.

The CAI method is an easy and inexpensive procedure, and is specially suited for Palaeozoic carbonate rocks, where other thermal maturity indices, such as the coal rank, vitrinite reflectance and Thermal Alteration Index (TAI), fail due to their biostratigraphic span, inadequate lithology, or their effective temperature range. Palaeo-temperature data derived from CAI values are frequently employed in basin analysis, metamorphic studies, and in determining thermal

aureoles related to short-term heating events (e.g., Kovács and Árkai, 1987; Nowlan and Barnes, 1987; Burnett, 1987, 1988, Rasmussen and Smith, 2001; Wiederer et al., 2002; Zhang and Barnes, 2007; Voldman et al., 2009). However, the CAI method is based on the assumption that heating events take place at constant temperature, which can conduct to inaccurate interpretations of palaeotemperatures or geothermal palaeogradients, particularly when dealing with complex and prolonged thermal histories. An accessible solution to this problem is proposed in this paper.

2. The conodont Color Alteration Index (CAI)

Conventionally, CAI values are determined under incident light on the outer basal margins and finest sections of the conodont elements by direct comparison with a set of laboratory produced standards or with the Munsell soil color chart (Epstein et al., 1977). Unaltered conodonts present a pale yellow and a smooth surface with silky brightness (CAI 1). Gradually increasing temperature results in successive carbonization processes of conodont elements that outcome in the color sequence light through dark brown (CAI 1.5–4) to black (CAI 5). Subsequent color changes towards grey (CAI 6), white (CAI 7) and finally translucent (CAI 8) are consequences of oxidation of organic matter, release of constitutional water and recrystallization. Epstein et al. (1977) and Rejebian et al. (1987) calibrated the different CAI stages, ranging in temperature from 50 to >600 °C, by means of laboratory experiments and extrapolated their experimental data to geologic time scales through the Arrhenius plot (Fig. 1). From these pioneer works, much research has been accomplished in order to quantify the conodont color alteration index, employing electron spin

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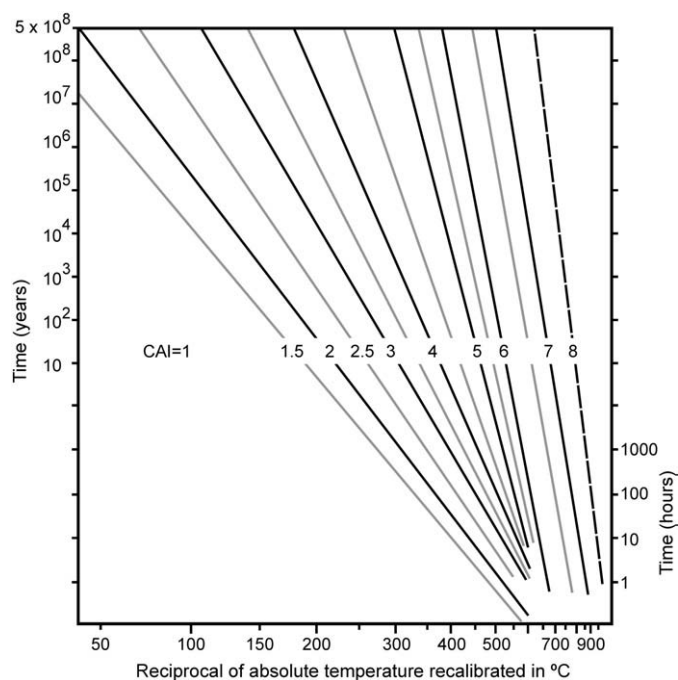


Fig. 1. Arrhenius plot of fields of CAI 1 through 8 from Epstein et al. (1977) and Rejebian et al. (1987). The gray curves are intermediate CAI values, usually discernable under the binocular microscope. Each CAI curve represents all combinations of temperature and time (isothermal conditions) that result in the corresponding CAI value.

resonance (Belka et al., 1987), organic geochemistry (Bustin et al., 1992; Marshall et al., 1999), spectral reflectance (Deaton et al., 1996), and color image analysis (Helsen et al., 1995; Bábek et al., 2008), among other techniques, with varying degrees of certainty. A general correlation between illite crystallinity and CAI values determined that the CAI range from 4 to 5.5 reflects anchizone boundaries.

The conodont color alteration sequence follows the Arrhenius reaction, therefore it is progressive, cumulative and irreversible. Short-term heating events do not necessarily produce the normal gradational conodont color alteration sequence. Usually, contact metamorphosed conodont elements have a broader range of CAI values, both locally from sample to sample and even within same sample. Moreover, contact metamorphosed conodonts are generally not deformed but strongly recrystallized: the size of single crystals on conodont elements suggests a close relationship between the growth of apatite crystals and the determined CAI value (Burnett, 1988; Königshof, 1992). Conodont elements recovered from hydrothermally altered rocks may show similar CAI distributions, but they are generally corroded and present a superficial grey patina (Rejebian et al., 1987). Under these circumstances, CAI values from 6 to 8 can be misleading in assessing temperatures, as the conodont color could result equally from oxidation of organic matter or from the temperature range experienced by the host rock during hydrothermal activity.

Besides thermal maturation, other factors such as dolomitization and diagenesis that could affect the CAI are usually accompanied by indicative textural alterations of the conodont surfaces, like patinas or coarse recrystallization (e.g., Burnett, 1988; Königshof, 2003; Voldman et al., 2008). Alternatively, robust conodont elements are usually darker, but this aspect has limited significance if similar forms are consistently selected for the color analysis. The influence of tectonics on CAI values is directly related to the variation of overburden levels, making it possible to use conodonts as geothermometers within orogenic belts (e.g., Rasmussen and Smith, 2001), though the combination of water and confining pressures may eventually lead to CAI suppression by inhibiting the charring processes, as it is observed in overpressured rocks (Epstein et al., 1977; cf. Hao et al. 2007). This mechanism is also invoked to explain oil production at

anomalously high temperatures within overpressured systems, where the organic matter maturation has been retarded. Under increasing conditions of regional dynamothermal metamorphism, where besides temperature, fluid pressure and oriented stress can play significant roles, recrystallization and ductile deformation of conodonts could be accelerated, in accordance with the metamorphism and ductile deformation experienced by the host rock (Teichmüller, 1987; Sudar and Kovács, 2006). Ultimately, the CAI can be influenced by the composition of the host-rock due to infiltrating of hydrocarbons or pyrite in the laminated conodont structure under reducing conditions or by enhanced thermal maturation related to radioactive decay of uranium (e.g., Helsen, 1997). This phenomenon usually represents less than 0.5 CAI units and can be avoided by working with similar lithologic types, especially with unweathered calcareous rocks.

CAI values caused by thermal alteration (e.g., sedimentary burial or contact metamorphism) can be predicted from a time–temperature Arrhenius graph, which shows the resulting CAI values produced after heating a conodont sample at constant temperature for a given period of time (Epstein et al., 1977; Rejebian et al., 1987). However, the application of this plot to complex geological histories is difficult because under natural conditions the heating occurs at variable temperatures, hampering the precise determination of palaeotemperatures or geothermal palaeogradients from CAI data. To overcome this problem, García López et al. (2001) proposed a graphic solution to estimate reliable palaeogradients from CAI data if the mean sedimentation rates and the chronostratigraphic boundaries of the units are known. Nevertheless, their method has some limits: (1) it evaluates the time the rock was above a specific temperature, despite the fact that CAI depends on the integral of temperature with time, (2) it assumes that the heating time is equal to the cooling time, oversimplifying the burial history, and (3) it is rather difficult to implement in a computer program. Herein, we present a simple model for the Arrhenius reaction of the conodont color alteration index (CAI), named to as EasyCAI, that overcomes all of these problems.

3. CAI calculations for complex thermal histories with variable heating rates

We assume that the chemical reactions considered here can be described by a pseudo-first order law,

$$dw/dt = -kw, \tag{1}$$

where w is the reactant concentration and k is the pseudo-first order reaction rate constant. The temperature dependence of k is expressed by the Arrhenius equation:

$$k = A \exp(-E_a/RT), \tag{2}$$

where E_a is the activation energy, A the pre-exponential factor, T the temperature and R the universal gas constant. Inserting this equation into Eq. (1) and solving it for constant T gives:

$$w = w_0 \exp\left(-Ae^{(-E_a/RT)}t\right) \tag{3}$$

We assume a linear dependence of the transformation ratio F of the reaction (ranging between 0 and 1) with CAI,

$$CAI = CAI_{\min} + w_F F \tag{4}$$

where $F = 1 - w/w_0$ and w_F is a constant weight factor. Combining Eqs. (3) and (4) results in:

$$CAI = CAI_{\min} + w_F \left[1 - \exp\left(-Ae^{(-E_a/RT)}t\right)\right], \tag{5}$$

This equation can satisfactorily model the time–temperature behaviour of any CAI curve (CAI_i) shown in Fig. 1, but does not explain the complete time–temperature dependence of the CAI, because each curve has a different slope and y-intercept. To overcome this problem, we assume a sum of multiple parallel reactions:

$$CAI = CAI_{\min} + \sum_i w_{F,i} \left[1 - \exp\left(-A_i e^{-Ea_i/RT} t\right) \right] \quad (6)$$

where the values of $w_{F,i}$, A_i , and Ea_i are adjustable parameters.

The major assumption of the CAI palaeothermometric method, as with other thermal maturation indexes conforming to the Arrhenius behaviour, is the extrapolation of these parameters to geological environments, provided that they are constant over a wide temperature range (Snowdon, 1979; Villa, 2001). For complex thermal histories, this uncertainty is downplayed by comparing CAI data with palaeotemperatures estimates from other indexes (e.g., illite crystallinity, vitrinite reflectance), in order to test the consistency of the results (see Brime et al., 2008 and references therein).

Under natural conditions, the thermal alteration of conodonts is neither isothermal nor as such fast reactions that they proceed instantly to completion upon achieve a particular temperature. Because of maturation effects on the organic material are additive and irreversible, the total maturity for a given stratigraphic level results from the sum of the maturities acquired in each successive time–temperature interval. For time-dependent temperatures histories, typical of geologic problems, the resultant maturity reflects the cumulative effects of thermal alteration as a conodont undergoes slow or rapid changes in temperature throughout its burial history.

The current approach considers that the conodont CAI depends on the time–temperature history in the same way as chemical reactions,

$$\int_{w_0}^w dw' / w' = - \int_0^t k(t') / dt' \quad (7)$$

Consequently, for non-constant temperatures, the resultant CAI value is governed by:

$$CAI = CAI_{\min} + \sum_i w_{F,i} \left[1 - \exp\left(-A_i \int_0^t e^{-Ea_i/RT(t')} dt'\right) \right] \quad (8)$$

In order to obtain Ea_i , A_i , and $w_{F,i}$ directly from the Arrhenius plot (Fig. 1) without employing an optimization procedure, we assume a constant weight factor ($w_{F,i} \equiv w_F$) and 12 parallel reactions (CAI_i), each of them corresponding to the CAI curves from Fig. 1. Additionally, we consider that for a given CAI_i, the *i*th reaction is at 50% of completion, while all the *j*th reactions with *j*<*i* are at 100% and all the *k*th reactions with *k*>*i* are at 0% of completion. For example, when the CAI = 5 is reached, its associated reaction is at 50% of completion. Meanwhile, all reactions corresponding to the lower CAI values are completed and all reactions corresponding to higher CAI values have not yet started. Under these conditions, the values of CAI_{min} and w_F are determined by:

$$\left[CAI_i - CAI_{\min} - w_F \sum_{j=1}^{i-1} (1-0) - w_F \sum_{k=i+1}^N (1-1) \right] / w_F \quad (9)$$

$$= 1 - \exp\left(-A_i e^{-Ea_i/RT_i}\right) = 0.5$$

In our case, as $N = 12$, CAI_i = {1.5, 2, 2.5, 3, 3.5, 4, 4.5, 5, 5.5, 6, 6.5, and 7}, and we are considering a 50% of completion of each *i*th reaction, the values of w_F and CAI_{min} must be 0.5 and 1.25 respectively. After rearrangement Eq. (9) yields:

$$\ln t = \frac{Ea_i}{R} \frac{1}{T} + \ln\left(\frac{\ln 2}{A_i}\right), \quad (10)$$

Table 1

Parameters of Eq. (8) obtained from the experimental extrapolation data of Epstein et al. (1977) and Rejebian et al. (1987) with CAI_{min} = 1.25, $w_F = 0.5$ and $N = 12$.

CAI _i	A _i (1/years)	Ea _i /R (K)
1.5	3.71868E11	13623.0311
2.0	7.98173E11	14925.7241
2.5	4.15231E12	17147.2317
3.0	2.16022E13	19368.7393
3.5	3.12823E14	22188.1258
4.0	1.44958E16	26067.4251
4.5	1.82228E19	32769.4825
5.0	4.91746E24	44318.0423
5.5	3.03821E27	51334.4557
6.0	5.01935E30	59864.7570
6.5	5.76012E29	64379.3110
7.0	6.96067E29	70092.2014

for a given CAI_i value. This way, the activation energies are given by the slopes of the Arrhenius curves and the pre-exponential factors by their y-intercept (Fig. 1, Table 1). The increment of E_a and A towards the higher CAI values is consistent with the thermal alteration progress, from dominantly carbonization to oxidation and recrystallization of the conodonts.

Our model applies to values of CAI from 1.5 to 7 and can be employed for time and temperatures scales ranging from those in the laboratory to all those of geologic interest, including brief igneous intrusions. The comparison between the proposed model and the Arrhenius plots of Epstein et al. (1977) and Rejebian et al. (1987) provided a standard deviation of 0.03 after testing 425 points over the conodont CAI curves.

In order to evaluate the theoretical CAI for a rock throughout its thermal history, we provided an Excel spreadsheet downloadable from the supplementary data site, called EasyCAI, that numerically integrates Eq. (8) with the parameters of Table 1. Using EasyCAI, a profile of CAI versus time can be computed for a given stratigraphic level if the time–temperature history is known or estimated.

4. Examples of model applications

The thermal degradation of conodonts is intimately related to the tectonic evolution of the sedimentary basins, as heat flow and subsidence vary through time in response to the lithospheric processes (Beardsmore and Cull, 2001; Allen and Allen, 2005). In a first study we apply EasyCAI on two contrasting thermal scenarios that share the total cumulative time and the last temperature acquired by the conodont sample (Table 2). Case 1 represents an isothermal history, following the graphic procedure of Epstein et al. (1977), while case 2 involves a nonlinear, more realistic, basin evolution. The results from both cases differ in ~0.5 CAI, which is often within the resolution of the visual CAI technique. However, as the exposure time to heat increase, the highest temperature experienced by the conodont sample governs its color alteration.

The accuracy of EasyCAI can be demonstrated by applying it to basins that have simple, well-defined thermal histories. In particular,

Table 2

Two hypothetical cases of conodont thermal alteration.

Time step	Temp. (°C)	Time step	Time	EasyCAI
<i>Case 1</i>				
1	150	0	0	1.25
2	150	400 Ma	400 Ma	3.69
<i>Case 2</i>				
1	20	0	0	1.25
2	60	140 Ma	140 Ma	2.00
3	80	220 Ma	360 Ma	2.36
4	150	40 Ma	400 Ma	3.14

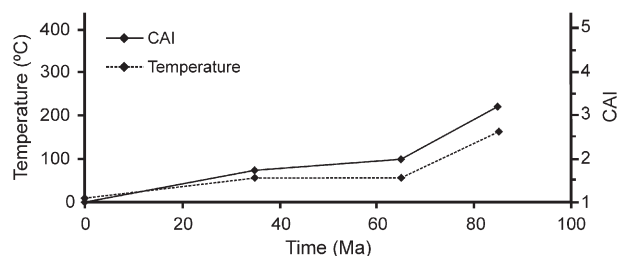


Fig. 2. Time-temperature history for the Cantabrian Zone and corresponding computed CAI values (data from García López et al., 2001).

foreland basins are ideal because they are characterized by uniform heat flow through time, as predicted by the lithospheric flexure models (e.g., Beaumont, 1981). An adequate example is provided by García López et al. (2001), who analysed the thermal alteration of Devonian conodonts from the external zone of the Variscan belt in NW Spain (Cantabrian Zone). According to their set of data, the regional CAI is 3.2 and the mean sedimentation rates are 35.0 m/Ma during the Devonian (thickness 1225 m), 0.8 m/Ma during the Tournaisian–Visean (25 m), and about 140.4 m/Ma during the Namurian–Westphalian (2800 m). After testing temperatures corresponding to several geothermal gradients, we matched the regional CAI with a gradient of ~35 °C/km, similar to the one predicted by the graphical method of García López et al. (2001) (Fig. 2). This value is also consistent with the geothermal gradient from the Appalachian foreland basin, where the CAI palaeothermometric technique was originally calibrated by Epstein et al. (1977).

An interesting example is provided by Kovács et al. (2006), who analysed the thermal alteration of conodonts related to a half-graben structure in southern Hungary. In the Mecsek half-graben area, middle Triassic conodonts were buried by the end of Bathonian by up to 4300 m of sediments, in contrast to a maximum of 90 m at the Villány Ridge area. Subsequent sedimentary burial took place during the Late Jurassic to Early Cretaceous, with maximum thicknesses of ~1000 m in both regions. According to Kovács et al. (2006), the middle Triassic conodonts from the Mecsek half-graben zone exhibit a CAI 4, whereas those from the Villány Ridge zone show a CAI 1, reflecting the great difference in burial depth. Nevertheless, when the thermal history of the Mecsek half-graben and the Villány Ridge areas are computed considering different palaeothermal gradients, it is observed that the CAI 1 suggests anomalously low palaeotemperatures (cf., Kovács et al., 2006) (Fig. 3). This is consistent with the studies of García López et al. (2001), who suggested to ignore the palaeotemperatures obtained from low CAI values (1, 1.5 or 2), as they yielded unrealistic low palaeothermal gradients in many localities of the Cantabrian Zone.

The thermal history of Ordovician conodonts from the Precordillera terrane of northwestern Argentina has also been constrained with EasyCAI. At the Don Braulio Creek, conodont-rich Middle Ordovician

Table 3 Thermal history model and corresponding computed CAI values for conodonts from the Don Braulio creek.

Time step	Burial history	Time step (Ma)	Thickness (m)	Estimated palaeotemperature (°C)	EasyCAI
1	Conodonts setting	0	0	20.00	1.25
2	Gualcamayo Formation (Middle–Upper Ordovician)	2	17	20.51	1.25
3	La Cantera Formation (Upper Ordovician)	5	142	24.77	1.26
4	Discontinuity	14	0	24.77	1.30
5	Don Braulio Formation (Upper late Ordovician)	6	56	26.45	1.31
6	Rinconada Formation (Silurian)	20	3750	138.95	2.81
7	Discontinuity	413	0	138.95	3.46
8	Neogene sedimentary deposits	2	200	144.95	3.46
9	Andean exhumation	3	–4165	20.00	3.46

carbonate platform sediments are covered by Upper Middle and Upper Ordovician foreland siliciclastic deposits, which are in turn covered by a thick Silurian mélangé (Astini et al., 1995). According to most supported hypothesis, the tectonic evolution of the Precordillera remained relatively stable until Andean triggering of the thin-skinned fold and thrust belt. Neogene synorogenic deposits outcropping in this sector comprise <100 m of stratigraphic thickness. In the present example, a 30 °C/km geothermal gradient results plausible to estimate the thermal history of the conodonts, taking into account the long-lasting geotectonic evolution of the Precordillera and its present Andean foreland position (Voldman et al., 2010). The thickness of the stratigraphic succession, the boundaries of the chronostratigraphic units and the calculated CAI values are given in Table 3.

Conodonts from the Don Braulio creek exhibit a CAI 3, in contrast to the calculated CAI value of ~3.5. The recorded value can be easily computed reducing the geothermal gradient to 20–25 °C/km. According to our results, the thermal alteration of those specimens involved quite variable heating rates, being primarily controlled by the deposition of the thick Silurian mélangé (Fig. 4). On the other hand, assuming that sedimentation would have continued in this sector of the Precordillera, without major interruptions, from the Ludlow to the end of Triassic times (as apparently occurred in other localities of the Precordillera), the integrated stratigraphic column would incorporate several thousand additional meters and the corresponding computed CAI values would be higher, reflecting the greater burial depths experienced by the conodont elements.

The temperature histories to which the CAI calculations were applied here are all in the diagenetic range; therefore, the

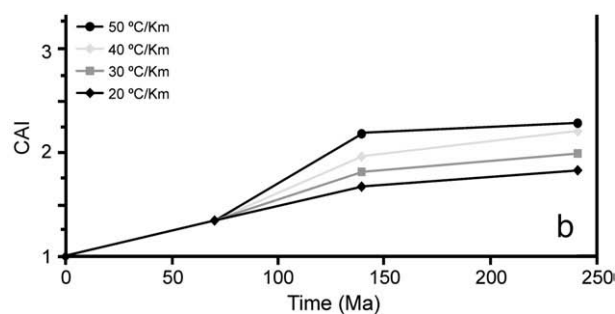
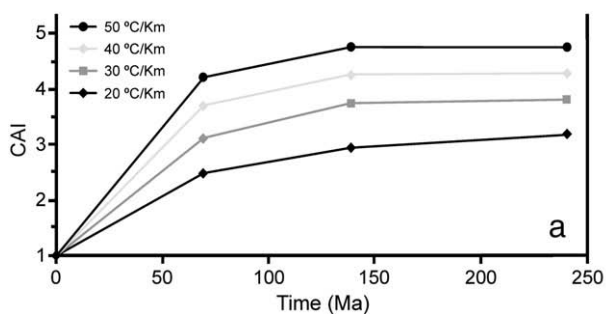


Fig. 3. Time-temperature histories with corresponding computed CAI values for (a) the Mecsek half-graben area and (b) the Villány Ridge area (data from Kovács et al., 2006).

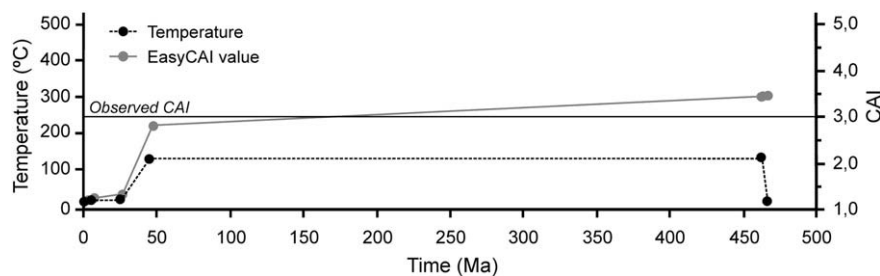


Fig. 4. Time–temperature history for conodonts from the Don Braulio Creek and corresponding computed CAI values.

temperature ranges where the higher activation energies contribute to the CAI were not reached. On the other hand, the results from EasyCAI can be compared with the results from the more popular vitrinite maturation models if vitrinite reflectance data are available (e.g., EASY%R_o from Sweeney and Burnham, 1990). This way, anomalous values given by one or another method can be recognised. More generally, it is possible to apply our approach to virtually any color index that follows an Arrhenius behavior (e.g., thermal alteration index of kerogens) in order to provide more accurate thermal analysis.

5. Summary and conclusions

The proposed Arrhenius reaction model, EasyCAI, considers the overall conodont maturation process as a series of parallel pseudo-first-order reactions. It can be easily implemented in a computer program to predict CAI values with changing time and temperature. EasyCAI precisely solves any type of thermal history: nondeposition, uplift and cooling, or heating rates that change with time. When applied to multiple stratigraphic levels, our equation can be used to compute profiles of CAI values with depth for comparison with borehole data and to optimize thermal history models; thus providing a new tool for quantitative CAI palaeothermometric analysis, and the interpretation of the thermal history of a basin.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.coal.2010.02.001.

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