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RESEARCH ARTICLE

Quantitative evaluation of organic richness from correlation of well logs and geochemical data: a case study of the Lower Permian Taiyuan shales in the southern North China Basin

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Abstract Marine-continental transitional shale is a potential energy component in China and is expected to be a realistic field in terms of increasing reserves and enhancing the natural gas production. However, the complex lithology, constantly changing depositional environment and lithofacies make the quantitative determination of the total organic carbon (TOC) suitable for marine shales not necessarily applicable to transitional shales. Thus, the identification of marine-continental transitional organic-rich shales and the mechanism of organic matter enrichment need to be further studied. As a typical representative of transitional shale, samples from Well MY-1 in the Taiyuan Formation in the southern North China Basin, were selected for TOC prediction using a combination of experimental organic geochemical data and well logging data including natural gamma-ray (GR), density (DEN), acoustic (AC), neutron (CNL) and U spectral gamma-ray (U), and TH spectral gamma-ray (TH). The correlation coefficient, coefficient of determination, standard deviation, mean squared error (MSE) and root mean squared error (RMSE) were selected to conduct the error analysis of the evaluation of different well log-based prediction methods, involving U spectral gamma logging, $\Delta\log R$, and multivariate fitting methods to obtain the optimal TOC prediction method for the Taiyuan transitional shale. The plots of TOC versus the remaining volatile hydrocarbon content and the generation potential from Rock Eval show good to excellent potentials for hydrocarbon generation. The integrated results obtained from the various log-based TOC estimation methods indicate that, the multivariate fitting method of GR-U-DEN-CNL combination is preferable, with the correlation coefficients of 0.78 and 0.97 for the entire and objective interval of the Taiyuan Formation respectively, and with the minimum MSE and RMSE values. Specifically, the U spectral gamma logging method based on single logging parameter is also a better choice for TOC prediction of the high-quality intervals. This study provides a reference for the exploration and development of unconventional shale gas such as transitional shale gas.

Keywords marine-continental transitional shales, total organic carbon, thermal maturity, well logs

1 Introduction

Shale gas is a clean, high-efficiency energy source, which occurs in organic-rich mudstone intervals (Zhang et al., 2004; Yan et al., 2009; Zhang et al., 2020). One of the most important parameters for shale reservoir evaluation is the calculation of the total organic carbon content (TOC), and it is also an important indicator for source rock evaluation (Fertl and Chilingar, 1988; Kamali and Mirshady, 2004; Sachsenhofer et al., 2010; Xi et al., 2020). Generally, TOC is used to evaluate the degree of organic matter enrichment in shale gas reservoirs. The richer the organic matter in the shale reservoirs, the greater the gas content and the better the reservoir quality. Therefore, quantitative determination of organic carbon content is the key to shale gas reservoir evaluation (Nie et al., 2017; He and Li, 2019). The TOC can be measured directly through laboratory experiments, and each sample is generally taken at a certain distance in the drilling process, which results in the discontinuity of the obtained TOC values (Wang et al., 2014). The use of these discrete TOC values to evaluate the entire source rock interval produces larger errors and affects the accuracy of the source rock evaluation results. Thus, the

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limited core analysis using traditional organic geochemical analyses has had difficulty meeting the demands of fine exploration in recent years. A solution to this problem is the continuous, high resolution information obtained from well logs (Zhao et al., 2016; Wang et al., 2018; Aziz et al., 2020). The common approaches to predicting the TOC based on well logs are the single parameter fitting method using the density log, natural gamma-ray log and spectral gamma-ray log, as well as the composite well logs including the $\Delta\log R$ and multivariate fitting methods (Hu et al., 2015; Bie et al., 2016; Wang et al., 2016a; Kenomore et al., 2017). Many scholars are dedicated to developing a reliable method of estimating the TOC from logging data in shale gas basins (Bie et al., 2016; Jiang et al., 2018; Chen et al., 2019; Godfray and Seetharamaiah, 2019; Harris et al., 2019; Jiang et al., 2019; Li et al., 2019c; Zhu et al., 2019; Aziz et al., 2020; Wang et al., 2020; Tan et al., 2021). In addition, with the rapid development of artificial intelligence technology in the field of oil and gas exploration, artificial neural networks, neurofuzzy, support vector machines and other techniques have also been proposed and applied to TOC prediction (Tan et al., 2013; Ouadfeul and Aliouane, 2015; Shi et al., 2016; Jiang et al., 2018; Rui et al., 2019; Liu et al., 2021). Artificial intelligence technology is more advanced than simple regression techniques or statistical correlations, but it is not applicable due to the need for large data sets and complicated regression computations.

Marine source rock formations have relatively high organic matter background values, and the amplitude difference between the reverse superimposed acoustic time difference curve and the resistivity curve is more obvious. Good results have been achieved using the traditional $\Delta\log R$ method in foreign marine source rock TOC predictions in a single well (Passey et al. 1990; Yu et al., 2017; Zhao et al., 2017). However, the geological and structural conditions in China are complex, resulting in the development of marine-continental transitional shale formations during multi-cycle structural evolution processes (Zhang et al., 2009; Zou et al., 2010; Shan et al., 2017; Cui et al., 2019; Li et al., 2019a, 2019b). There are often sedimentary facies transitions in lateral, and limestone and sandstone interlayers often occur in the vertical direction, resulting in different changes in the thickness, distribution, TOC, the mineral and rock composition of gas-bearing shales. These factors change the hydrocarbon-generating capacity, which in turn introduces some uncertainty into the evaluation of the source rocks. Therefore, TOC prediction technology that works well for marine shale reservoirs may not be applicable to marine-continental transitional shales.

Earlier studies have shown that as a typical representative of marine-continental transitional shale in China, the shale source rocks of the Taiyuan Formation in the Zhongmu Block, southern North China Basin, have a good gas generation potential and a well fracturing development zone (Li et al., 2016; Tang et al., 2016, 2020; Zhou et al., 2016). Well MY-1 in the Zhongmu Block is the first exploration well for marine-continental transitional shale gas in the southern North China Basin. In this study, both single and composite logging-based TOC prediction methods were used on the shale sample from Well MY-1 in the Taiyuan Formation to identify a reliable method of TOC estimation via correlation of predicted TOC values with the measured TOC values. These research results not only have guiding significance for the exploration and development of transitional shale gas in the Zhongmu Block in the southern North China Basin, but also effectively aid in the exploration and development of unconventional oil and gas in other areas.

2 Materials and methods

2.1 Study area and sampling

The Zhongmu Block is located to the east of Zhengzhou City, Henan Province. The landform is a plain covered area characterized by a relatively stable structure and a gentle stratum occurrence (Fig. 1). Its tectonic evolution has successively included the development of a littoral-neritic subsidence basin during the Cambrian-Middle Ordovician, craton paleo-land uplift and denudation period during the Late Ordovician-Devonian, the development period of a marine-continental transitional subsidence basin during the Carboniferous-Permian, and the development of Triassic-Quaternary continental basins (Qiu et al., 2018). The evaluation result of shale gas resource potential in China has revealed that the Carboniferous-Permian transitional shales of the Taiyuan Formation and Shanxi Formation in this block have good exploration prospects (Li et al., 2016; Zhou et al., 2016). Among them, the Lower Permian Taiyuan Formation is generally composed of three sets of limestone and two sets of black mudstone containing coal seams. The overall performance is a transgressive retrograding sequence, in which the shale section is a lagoon facies and the limestone section is a restricted platform facies. The lower part of the Shanxi Formation is mainly sandstone, and the upper part is generally comprised of black mudstone and coal seams. The overall performance is a regressive prograding sequence. The fact that the barrier-lagoon sedimentary system is the main sedimentary environment for the development of high-quality shale reveals the excellent hydrocarbon generation potential of Taiyuan Formation shale. This is consistent with the results of a previous study, in which, samples from Well MY-1 in the Lower Permian Shanxi and Taiyuan Formations shales in the Zhongmu Block were characterized based on their geochemical, mineralogical, petrological, petrophysical, and pore microstructural characteristics using a variety of techniques (Tang et al., 2016). Through the comprehensive evaluation of multiple criteria (Charpentier and Cook,

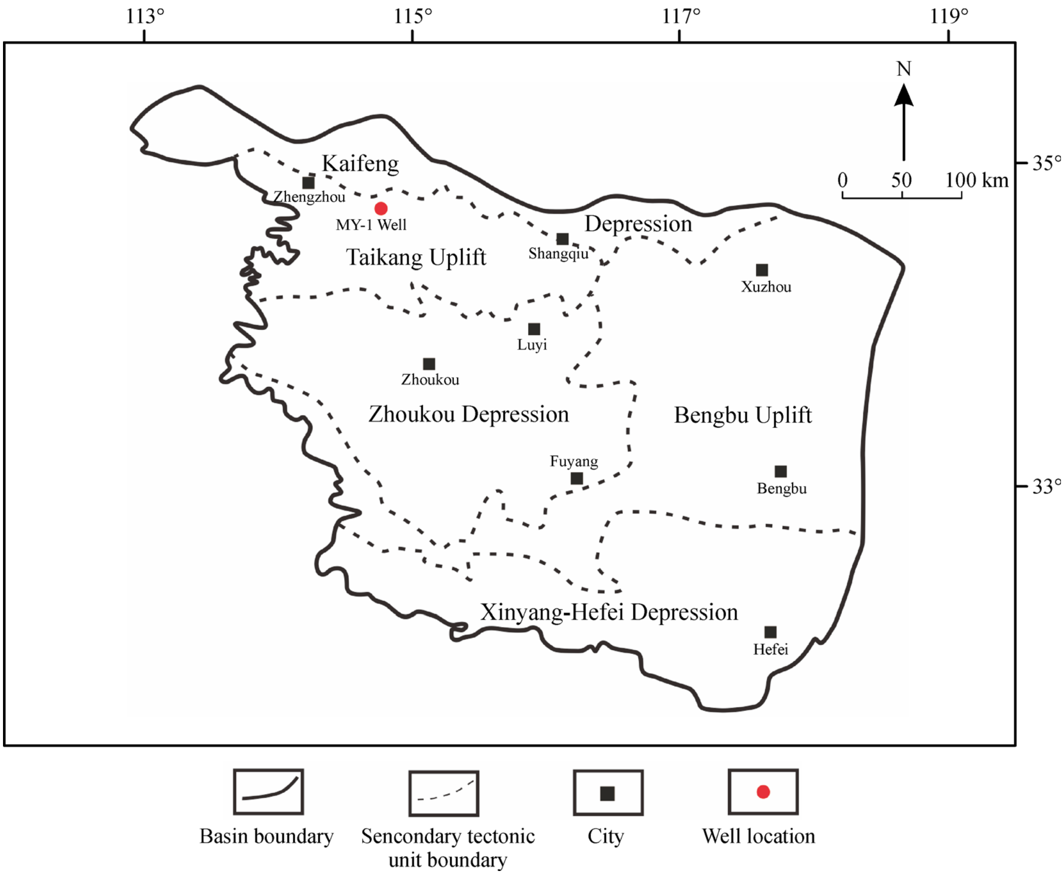

Fig. 1 Simplified structural map of the southern North China Basin, showing well locations (modified from Sun, 1996, with permission from the Chinese Journal of *Petroleum Exploration and Development*).

2011; Gross et al., 2015), the following conclusions have been drawn. At a depth of 2930–2952 m, the Taiyuan shale is the potential candidate for shale-gas prospecting studies because of its larger thickness, higher thermal maturity, good porosity, and ideal geochemical characteristics including TOC, S1 + S2, good brittleness, and lower TH/U values (Tang et al., 2016, 2020). These advantages make it the most desired development layers between the two red dashed lines shown in Fig. 2.

TOC predications based on well logs are commonly utilized due to its continuous data availability throughout the entire zone of interest (Harris et al., 2019; Aziz et al., 2020). To further study the correlations between the organic geochemical characteristics and the logging data, a complete suite of well log data for Well MY-1 was obtained in this study. Moreover, a total of 17 core samples from the Lower Permian Taiyuan Formation shale were collected from Well MY-1 at depths of 2895.51–2963.00 m, which were used to evaluate the quantity, quality, and type of the organic matter present in the Taiyuan Formation. It should be noted that, a total of 8 samples within the candidate interval of 2930–2952 m were analyzed emphatically because of their good gas potential.

2.2 Organic geochemical analyses

The TOC content analysis was completed using a CS-230 carbon/sulfur analyzer (Leco, USA). The analysis procedure was as follows in accordance with Chinese standard GB/T 19145-2003 (GB, 2003). First, 100 mg of shale samples were first crushed to powder below 100 mesh (< 150 mm) in particle size. Then, the powder were placed in a crucible and infused with 5% dilute hydrochloric acid at 80°C for 24 h to remove any carbonate minerals. Deionized water was used to rinse the sample 6 times to remove any residual dilute hydrochloric acid solution. The acid-treated sample was dried under vacuum at 80°C for 24 h. The dried powder sample was mixed with a combustion promoter composed of iron powder and tungsten-iron alloy. Oxygen was used as the combustion gas, and nitrogen used as the carrier gas. The combustion temperature of the automatic analyzer was increased to 3000°C, and the TOC content was calculated based on the area of the carbon dioxide peak produced by the combustion of the organic matter.

Rock-Eval pyrolysis is commonly used to obtain the key parameters used to assess the amount and type of the organic matter, as well as the maturity level of the organic

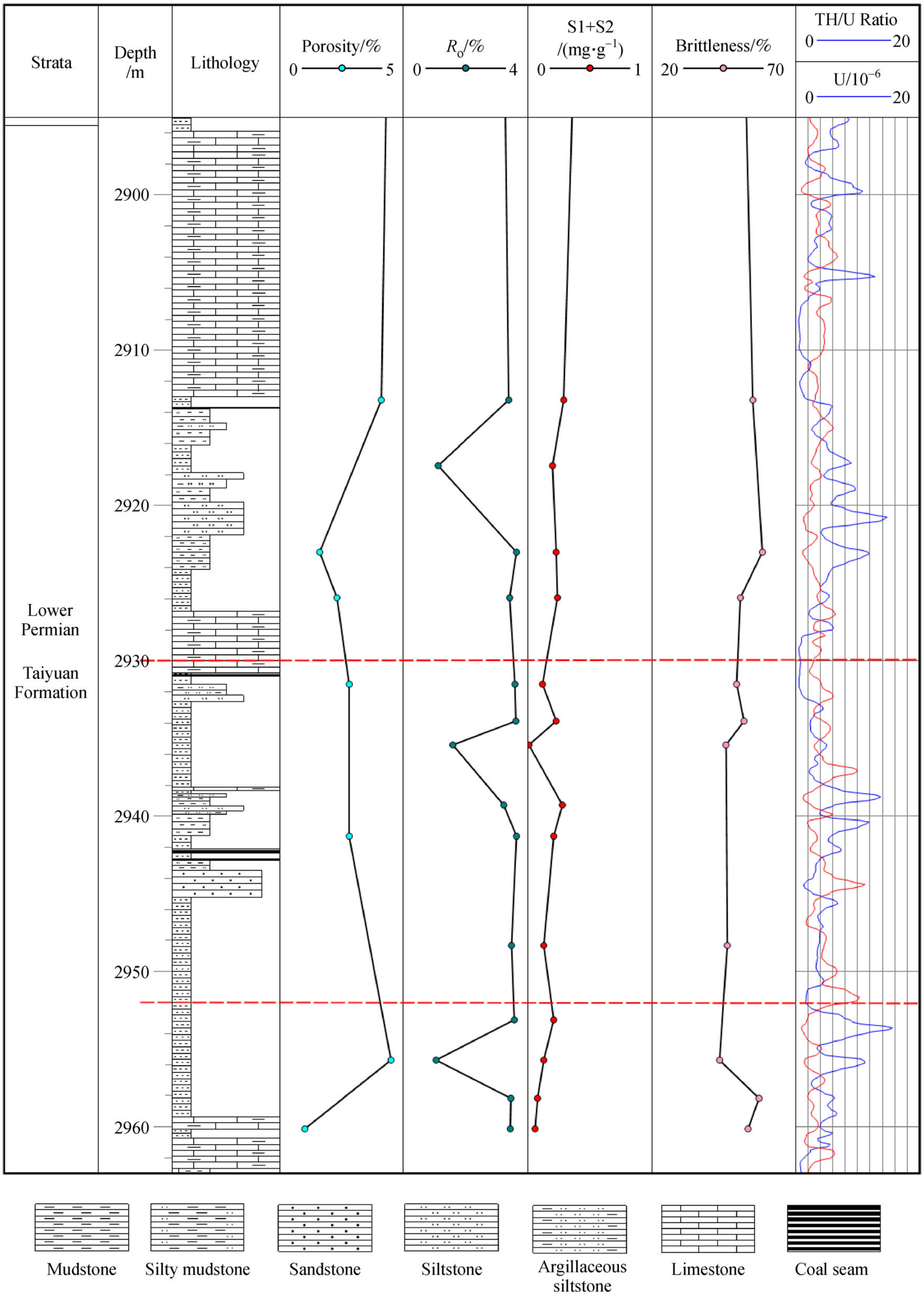

Fig. 2 Stratigraphic column showing the porosity, vitrinite reflectance, gas potential, brittleness, and spectral gamma-ray log data for the Lower Permian Taiyuan Formation (modified from Tang et al., 2016, with permission from Elsevier).

matter in shale rocks (Dong et al., 2020). The sample preparation process for the Rock-Eval pyrolysis was the same as that for the TOC measurements. The experimental procedure was conducted using a Rock-Eval 6 instrument and a minimal amount (80 mg) of crushed rock sample. The temperature-rising program for the sample analysis was as follows according to Chinese Standard GB/T 18602-2012 (GB, 2012). First, the temperature was kept at 300°C for 3 min to measure the volatile hydrocarbon S1 content (S1); and then, the temperature was increased to 600°C at a rate of 25°C/min to determine the remaining hydrocarbon generative potential (S2) (Jarvie and Lundell, 1991; Jarvie et al., 2001). In addition, to determine the level of organic metamorphism (LOM), the vitrinite reflectance (R_o) values of 7 samples were also measured using an MPV-SP microphotometer under oil immersion according to Chinese Standard SY/T5124-1995 (SY, 1995).

2.3 TOC estimation from logging data

Laboratory analysis of the core and cutting samples is the basic method used to obtain the TOC content of the source rocks (Zhu et al., 2003; Jarvie, 2015; Zhao et al., 2017; Wang et al., 2019a). However, owing to the difficulty of sampling, it is nearly impossible to obtain continuous and complete TOC content changes and distribution characteristics in source rock profiles. Since Beers (1945) and Swanson (1960) discovered the relationship between the TOC content and rock radioactivity, the method of fitting the TOC content using well logging data has attracted increasing amounts of attention due to its available information with high resolution and continuity (Guo et al., 2009).

2.3.1 Single parameter fitting method

The increase in the organic matter content can be reflected in the relative changes of multiple parameters along the logging curve (Meng, 2018; He and Li, 2019). Conventional logs such as density, resistivity, neutron, natural gamma-ray, spectral gamma-ray, and acoustic logs show abnormal responses because of the presence of TOC in the encountered rock unit (He et al., 2016; Wang et al., 2019a). Such a response can be further utilized to recognize and estimate the source rock potential. In the study area, the core data can be used to scale to the corresponding logging data, to draw a corresponding intersection diagram, and then to establish a single-factor model of the TOC content related to logging data (Aziz et al., 2020). Finally, the mathematical relationships are developed and commonly used to estimate and the predict log-based TOC.

2.3.2 $\Delta\log R$ method

The $\Delta\log R$ method is an available TOC calculation method involving a rigorous petrophysical model derivation related to the logging curve and maturity, which was proposed by Passey et al. in 1990 based on Archie's formula. This method can be used to obtain continuous TOC content in depth employing overlapping curves, in order to compensate for the discontinuity and the limited availability of core/well cutting geochemical data (Passey et al., 1990; Liu et al., 2011; Gao et al., 2012). When the content of organic matter increases, the acoustic logging value and the corresponding resistivity value increase. On this basis, the reverse superposition of the acoustic logging and resistivity (usually deep resistivity) logging curves can be used to indicate the organic matter enrichment. Taking the fine-grained non-source rock as the baseline corresponding to the overlap of the two curves, the amplitude difference between the two curves is defined as $\Delta\log R$ (Eq. (1)). This method uses the difference between the logging curve value and the shale baseline to reflect the organic matter content, which follows the principle that the greater the difference, the higher the TOC content. It works well in relatively pure shale zones, but it is inferior in formations containing low-resistivity minerals such as pyrite.

$$\Delta\log R = \log_{10}(R/R_{\text{baseline}}) + 0.02 \times (\Delta t - \Delta t_{\text{baseline}}), \quad (1)$$

where $\Delta\log R$ is the curve separation measured in the logarithmic resistivity cycle; R and Δt are the resistivity and the transit time, respectively; and R_{baseline}, $\Delta t_{\text{baseline}}$ are the resistivity and the transit time corresponding to the baseline (non-source rock), respectively. The units of each parameter are $\Omega\cdot$cm, μs$\cdot$ft^{-1}, respectively.

A certain relationship exists between the value of $\Delta\log R$ and the TOC content, and the coefficient is the maturity parameter, that is, the level of organic metamorphism (LOM). Passey et al. (1990) produced a diagram relating $\Delta\log R$ to TOC under different maturity levels through a large amount of data calculations. Using the maturity parameter, the $\Delta\log R$ amplitude difference is converted into the TOC value using Eq. (2).

$$\text{TOC} = (\Delta\log R) \times 10^{(2.297 - 0.1688\times\text{LOM})}, \quad (2)$$

In practical applications, the LOM index is usually converted into a function of the vitrinite reflectance (R_o), which is one of the most important parameters used to determine the maturity of organic matter (Zhu et al., 2019). Generally, the corresponding relationship between R_o and the organic matter maturity stage is: immature stage corresponds to R_o of 0 – 0.5%, low-mature stage corresponds to R_o of 0.5% – 0.7%, mature stage corresponds to R_o of 0.7% – 1.3%, and high-mature stage corresponds to R_o of 1.3% – 2.0%, the over-mature stage corresponds to R_o greater than 2.0%. Meng (2018) obtained the conversion relationship between R_o and LOM under different maturity stages as shown in Eq. (3) – Eq. (5).

The stage of low-maturity to maturity:

$$\mathrm{LOM} = 12.74\ \log_{10}R_o + 11.15. \tag{3}$$

The stage of maturity to high-maturity:

$$\mathrm{LOM} = 7.95\ \log_{10}R_o + 10.83. \tag{4}$$

The stage of high-maturity to over-maturity:

$$\mathrm{LOM} = 21.62\ \log_{10}R_o + 7.22. \tag{5}$$

2.3.3 Multivariate fitting method

It is feasible to select multiple well logs to establish a multiple linear regression equation to improve the prediction effect of the TOC content (Heidari et al., 2011; Wang et al., 2019b). The multiple linear regression method presupposes that Y is a random variable predicted by X_1, X_2,..., X_m. The existence of a certain linear relationship between them led to the establishment of the m-ary linear regression model by using Eq. (6) (Guo et al., 2009):

$$Y = \beta_0 + X_1\beta_1 + X_2\beta_2 + ... + X_m\beta_m + \xi, \tag{6}$$

where β_0, β_1,..., β_m are the solve-for parameters and ξ is random error.

Conventional logging data are the result of the random response of the formation to the minerals and fluids in the formation, and it is closely related to the TOC. Therefore, based on core experiments and logging data analysis, a multivariate linear fitting model (Eq. (7)) of the conventional logging data and TOC can be established by selecting the logging curves sensitive to the TOC response.

$$\mathrm{TOC} = a\cdot\mathrm{DEN} + b\cdot\mathrm{RT} + c\cdot\mathrm{CNL} + d\cdot\mathrm{GR} + e\cdot\mathrm{U} + f\cdot\mathrm{AC} + g, \tag{7}$$

where a, b, c,..., and g are the solve-for parameters which may be positive, negative, or zero; DEN, RT, CNL, GR, U, and AC represent the corresponding logging data on density, resistivity, neutron, natural gamma-ray, spectral gamma-ray, and acoustic, respectively. It should be noted that other logging data can also be considered according to the actual fitting results.

Combining the above methods to conduct a research on selected shale samples to obtain the optimized well log-based TOC prediction method, which is suitable for the transitional shale of Taiyuan Formation in Zhongmu Block (Fig. 3).

3 Results and discussion

3.1 Organic geochemical characteristics

The results of the geochemical experiment involving the Rock-Eval and maceral compositions of the shale samples from the Taiyuan Formation are presented in Table 1. The vitrinite reflectance (R_o) of the organic shale in the study area is mostly above 3.0%, with an average value of 3.5%. This indicates that the regional organic shale is in the over-mature stage, reaching the standard for generating dry gas (Tissot and Welte, 1984), and the shale formation has a large potential for gas generation. Previous research further supports this observation, which reported that an abnormally high thermal maturity (> 3.0%) occurred in this area, mainly caused by thermal events (Xu et al., 2005, 2011; Cheng et al., 2011; Zhao et al., 2011; Wu et al., 2015). The level of organic metamorphism (LOM) is one of the key parameters for the maturity evaluation of organic matter. LOM = 12 corresponds to the beginning of the organic matter over-mature stage (Passey et al., 1990).

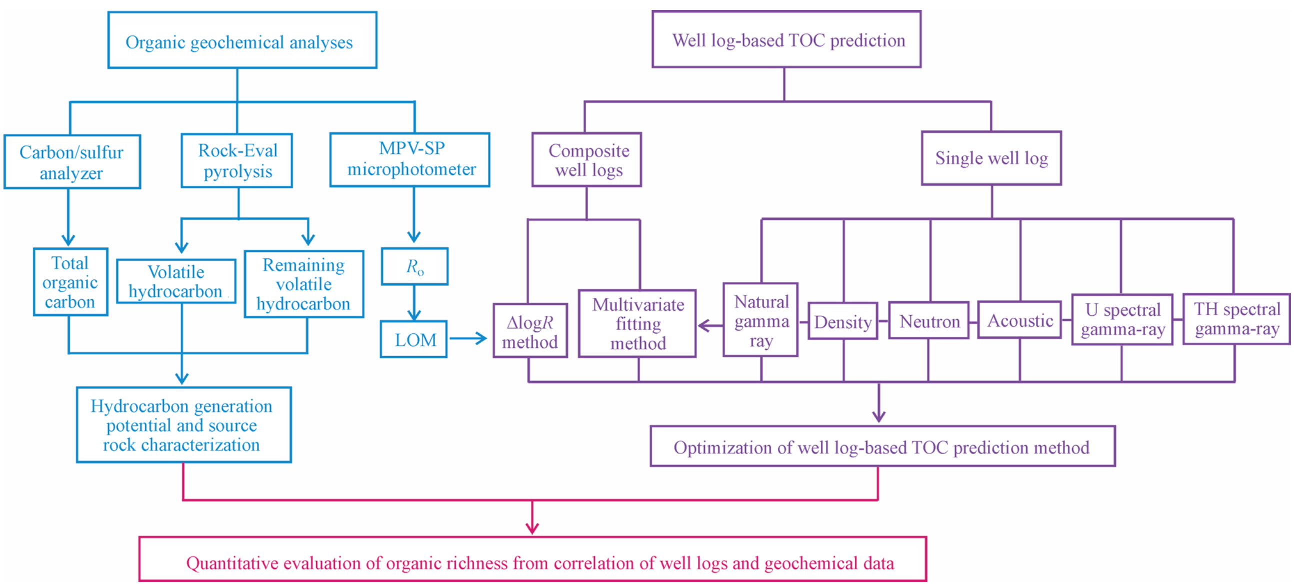

Fig. 3 Workflow adopted in this study.

Table 1 TOC, Rock-Eval, vitrinite reflectance and maceral composition data for the Taiyuan samples

Sample	TOC/%	Volatile hydrocarbon /($mg \cdot g^{-1}$)	Remaining volatile hydrocarbon /($mg \cdot g^{-1}$)	Vitrinite reflectance/%	LOM	Maceral composition			Type index	Kerogen type
						Liptinite/%	Vitrinite/%	Inertinite/%		
TY-1	3.74	0.05	0.24							
TY-2	0.92	0.04	0.16							
TY-3	1.87	0.04	0.19							
TY-4	1.93	0.03	0.21	3.58	19.19	0.0	7.5	92.5	−98	III
TY-5	1.55	0.06	0.54							
TY-6	1.75	0.02	0.1	3.59	19.22	5.9	11.7	82.4	−88	III
TY-7	4.14	0.02	0.21							
TY-8	1.33	0.04	0.13							
TY-9	1.69	0.07	0.21	3.56	19.14	0.0	16.1	83.9	−96	III
TY-10	2.25	0.03	0.18	3.5	18.98	34.3	6.9	58.8	−47	III
TY-11	2.45	0.03	0.19							
TY-12	5.06	0.03	0.14							
TY-13	1.67	0.03	0.18	3.46	18.87	0.0	15.3	84.7	−96	III
TY-14	1.88	0.03	0.1							
TY-15	2.55	0.02	0.06	3.48	18.93	0.0	17.5	82.5	−96	III
TY-16	1.89	0.03	0.03	3.34	18.54	0.0	36.8	63.2	−91	III
TY-17	1.96	0.04	0.28							

Since the organic shale in the study area is in the over-mature stage, Eq. (5) was used to calculate the LOM, which corresponds to the stage from high maturity to over-maturity, and the calculated average LOM is approximately 19. This further indicates the over-maturity of the organic matter.

The maceral components of the kerogen can be divided into four types: the sapropelinite group, the liptinite group, the vitrinite group, and the inertinite group. Among them, the sapropelinite group is mainly comprised of algal plastids, and amorphous bodies decomposed by aquatic organisms and algae. It belongs to Type I Sapropelic organic matter and has a high hydrocarbon generation potential. The liptinite group is dominated by cutinite, resinite and sporinite from higher plants. It is a typical type II organic matter and has a strong oil-generating ability. The vitrinite group and inertinite group consist of organic matter from higher plants, representing type III humic organic matter. The kerogen type can be determined according to the type index (TI) calculated from the maceral composition using Eq. (8). $TI \geqslant 80$, $80 > TI > 40$, $40 > TI > 0$, and $TI < 0$ indicate the organic matter of type I, type II, type II_2, and type III, respectively (Cao, 1985). The negative TI values in Table 1 correspond to the type III kerogen of the Taiyuan shale samples from the Well MY-1 (Tang et al., 2016).

$$\mathrm{TI} = \text{type index} = 100 \times (\text{sapropelinite}, \%) + 50 \times (\text{liptinite}, \%) - 75 \times (\text{vitrinite}, \%) - 100 \times (\text{inertinite}, \%). \quad (8)$$

The hydrocarbon generation potential and source rock characterization were also investigated using the geochemical data, such as the volatile hydrocarbon content (S1), remaining volatile hydrocarbon content (S2) and TOC content. The hydrocarbon generating potential and organic richness of the selected rock samples were evaluated using TOC (wt.%) content and S2 (mg/g). According to the criteria of Peters and Cassa (1994), the majority of the shale samples have a good to excellent source rock potential, and only a single sample has a fair source rock potential (Fig. 4).

The weight percentage of the TOC contents and the generation potentials (GPs) of the selected samples were plotted to explore the source rock characterization (Fig. 5). The GP is equal to the sum of the S1 and S2 values. TOC, S1, and S2 are the primary variables used to predict the richness and generation potential of the organic matter in a shale source formation (Wang et al., 2019a). The important information required in the initial exploration stages is the presence or absence of an effective source rock. In the study area, most of the samples plot in the good to excellent source rock potential region, except for one sample that plots in the fair region. The source rock characterization is consistent with the analysis of the hydrocarbon generating potential. In general, the Taiyuan samples are good source rocks with a good hydrocarbon generation potential.

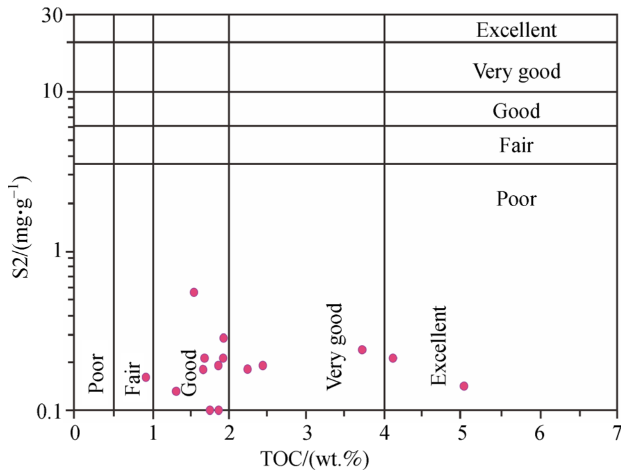

Fig. 4 Cross-plot of TOC versus remaining volatile hydrocarbon content showing the hydrocarbon generation potentials of selected Taiyuan samples.

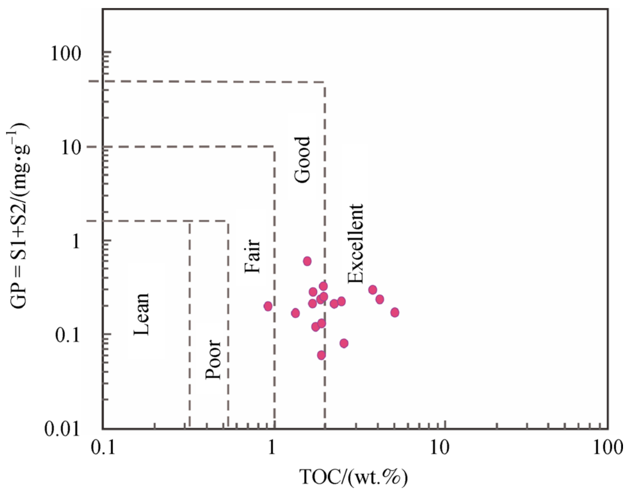

Fig. 5 Cross-plot of TOC versus generation potential showing the source rock characterization of selected Taiyuan samples.

3.2 Single parameter fitting method

The inherent theories of the various logging methods can better explain the response mechanism of shale gas reservoirs (Hao et al., 2012; Meng, 2018).

In natural gamma-ray (GR) logging, the gas-bearing shale formation has a high natural gamma value, reflecting the high radioactivity of the formation. The response mechanism is that, the shale layer is mainly composed of clay minerals, and it is rich in fine sandy and muddy particles with a large specific surface area and strong adsorption capacity for the radioactive elements of uranium (U), thorium (TH), and potassium (K), which leads to a high natural gamma value. Moreover, thorium and potassium can become trace elements that constitute clay minerals, which also leads to high values of natural gamma. In addition, shale deposition generally occurs in a strongly reducing environment with deep water bodies, and a low deposition energy is conducive to the enrichment of organic matter. Organic matter has a strong ability to adsorb radioactive elements similar to clay minerals, resulting in a high value of natural gamma.

In spectral gamma-ray logging, the high U value represents the sedimentary water environment of the shale, and it can better indicate the enrichment of organic matter (Adams and Weaver, 1958; Pan et al., 1990; Wang et al., 1990; Kochenov and Baturin, 2002; Johnston, 2004; Jarvie et al., 2007; Li, 2014; Liang et al., 2018). As a type of chemically stable radioactive element, TH is adsorbed by the organic matter and/or clay minerals in the shale area, and it mainly comes from debris, such as silt and shore erosion material, carried by terrestrial sources (Shi et al., 1996). A high TH value indicates a depositional environment close to the terrestrial source area. Therefore, in the response mechanism of the spectral GR logging for gas-bearing shale, the U curve emphasizes the level of the organic matter content in the shale layer, whereas the TH curve emphasizes the level of the shale content.

In density (DEN) logging, the pore space in the gas-bearing shale is dominated by natural gas, which causes the density of the gas-bearing shale interval to be lower than that of a conventional shale interval. Moreover, because the density of organic matter is much lower than that of minerals, the presence of organic matter will lead to a decrease in the value of the density logging curve corresponding to the organic-rich zone.

The shale formation has a high irreducible water saturation, which causes the compensation neutron logging (CNL) of the shale interval to exhibit a high value. Occasionally, the gas-bearing shale layer has an abnormally low value due to the “excavation” effect of the natural gas. Therefore, CNL is dominated by high values in gas-bearing shale intervals, and locally, it exhibits abnormally low values in the gas-saturated intervals.

In resistivity (RT) logging, shale has a better conductivity than sandstone and carbonate due to the large amount of micropores and open cracks developed in shale, which caused the formation water to penetrate the entire shale formation. Furthermore, the conductivity of gas-bearing shale is higher because the gas in the shale occupies the pores and the cracks affecting the conductivity, and the organic matter has a higher resistivity.

The acoustic (AC) time difference of gas-bearing shale appears to be high. One reason for this is that the pore-fracture spaces are saturated with gas, which shows down the speed of the acoustic wave during the propagation process, resulting in the phenomenon of “cycle-skip.” Another reason is the high organic matter content in gas-bearing shale, which has a low acoustic wave propagation speed, and increases the acoustic time difference (AC value).

As was previously mentioned, the increase in the organic matter content will be reflected by relative changes in multiple parameters of the logging curve. Taking 17 Taiyuan shale samples from Well MY-1 in the study area as an example, the TOC values of the 17 samples were analyzed via laboratory experiments. Single parameter fitting analysis was performed between the experimentally measured TOC values (TOC_EM) and the logging data including natural gamma-ray, density, acoustic, neutron, and spectral gamma-ray, at the corresponding depths (Table 2). It can be found that only the U spectrum gamma logging has a better fitting relationship with the measured TOC. In particular, 8 samples from the objective interval were selected for fitting analysis, and the fitting results of the U spectrum gamma logging are still the best. Moreover, compared with the entire Taiyuan interval, the correlation coefficient (R) is greatly improved to 0.91.

The linear regression fitting formulas for the TOC and gamma spectrum U curve for the entire interval (EI) and for the objective interval (OI) are expressed as following in Eq. (9) and Eq. (10).

Entire interval:

$$\mathrm{TOC} = 0.4422\mathrm{U} + 0.831 \qquad R^2 = 0.3062. \tag{9}$$

Objective interval:

$$\mathrm{TOC} = 1.22\mathrm{U} - 2.6899 \qquad R^2 = 0.835, \tag{10}$$

where TOC is the experimentally measured organic carbon content, %; and U is the reading of U spectral gamma logging at the corresponding depth, 10^{-6}.

The single parameter fitting results show that the U spectrum gamma logging has the best fitting effect for both the entire interval and the objective interval (Table 2). The Pearson's correlation coefficients (R) were 0.55 and 0.91 for the EI and OI respectively, followed by the GR (0.35 and 0.56) and TH (0.17 and 0.53). Based on the fitting equations, the corresponding relationships between the measured TOC value and the logging data are consistent with the conclusion summarized above, that is, TOC increases as the GR, AC, U, and TH values increase, and decreases as the DEN value increases. However, the measured TOC and CNL do not show good positive correlations, which may be due to the natural gas digging effect in the layers with a high gas saturation, causing local abnormally low values, and affecting the fitting effect.

In addition, the depositional environment of the shale reservoirs can be identified using spectral gamma logging by analyzing the TH, U, and K contents, and the TH/K and TH/U ratios. The analysis of the depositional environment of shale reservoirs is helpful in distinguishing whether the reservoir rock was deposited in a high-energy or low-energy environment, an oxidizing or reducing environment, and it has an important reference significance for the comprehensive evaluation of shale gas reservoirs (Tang et al., 2016).

3.3 Δlog*R* method

This theoretical approach requires that a baseline to be determined first. After this, the Δlog$R$ can be calculated from the baseline (Zhu et al., 2019). When using the Δlog$R$ method for TOC fitting, it is necessary to consider both the value of Δlog$R$ and the degree of thermal evolution. As can be seen from Table 1, the degree of thermal evolution of each sample is approximately the same, and the average LOM after rounding is 19. In both the EI and the OI in the Taiyuan Formation, the TOC_Δlog$R$ value calculated using the Δlog$R$ method has a poor match with the experimentally measured TOC_EM value (Figs. 6 and 7). The coefficients of determination (R^2) for the linear fitting of the two intervals are only 0.06 and 0.28 for the EI and the OI, respectively, and the predicted TOC value even appears to be negative. The prediction of the TOC content using the AC logging overlain with the RT logging does not produce a good estimation of the TOC content. The reasons for the poor effect of the Δlog$R$ method in predicting the TOC in the study area may mainly be as follows:

1) There may be errors in the estimation of the maturity parameter (LOM). The LOM index proposed by Passey et al. (1990) is generally an integer and the accuracy is not good.

2) The determination of the baseline is an important and difficult problem in Δlog$R$ method, which mainly involves the proper scaling of the two curves. The artificial selection of the baseline introduces a certain risk, and the selected baseline value is not unique, which affects the accuracy of the AC and RT values corresponding to the extracted mudstone baseline (Tan et al., 2021).

3) The traditional Δlog$R$ method was proposed based on marine and normally compacted source rock formations. When this method is applied to marine-continental transitional shale reservoirs, the terrestrial sedimentary

Table 2 Pearson's correlation analysis of relationships between TOC and logging data for single parameter fitting in the entire interval and objective interval of Taiyuan Formation

Interval	Correlation	GR	DEN	AC	CNL	U	TH
EI	Equation	$y = 0.0128x + 0.80$	$y = -1.2753x + 5.3707$	$y = 0.0059x + 1.8155$	$y = -0.004x + 2.3602$	$y = 0.4422x + 0.831$	$y = 0.0319x + 1.7995$
	R	0.35	0.28	0.07	0.02	0.55	0.17
OI	Equation	$y = 0.0198x + 0.2157$	$y = -0.6512x + 4.0244$	$y = 0.0104x + 1.6991$	$y = -0.0187x + 3.0101$	$y = 1.22x - 2.6899$	$y = 0.0943x + 1.277$
	R	0.56	0.09	0.12	0.09	0.91	0.53

formations have high conductive components, so there is no obvious abnormality in the resistivity curve, which leads to the occurrence of large errors.

4) The overly frequent lithology changes in the marine-continental transitional shale reservoirs, cause the heterogeneity of the minerals to be more complicated, and the thickness of the continuous pure mudstone layer does not exceed 15 m.

Generally, the background value of the organic matter in the marine source rock formation is relatively high, and the amplitude difference between the reverse superimposed AC curve and the RT curve is more obvious. The traditional $\Delta\log R$ method has achieved good results in TOC predictions of marine source rocks (Guo et al., 2012). However, terrestrial source rock formations are widespread in China, which results in problems such as low TOC background values, as well as unconspicuous amplitude difference between the AC curve and the RT curve (Hu et al., 2016). Owing to the differences in the geological factors, the application effect of the traditional $\Delta\log R$ method is not very good, especially for continental, marine-continental and high-over-mature marine shale reservoirs (Chen et al., 2015; Wang et al., 2016a; Qin et al., 2018). Consequently, a series of improved $\Delta\log R$ methods have been successively proposed according to the geological backgrounds of different research areas. Li et al. (2013) used the U curve and the $\Delta\log R$ combination method to calculate the TOC of the Lower Paleozoic continental shale in the Sichuan Basin, which fully considered the susceptibility of the continental shale to the lithology, resulting in the calculated value being closer to the measured value. Liu et al. (2014) developed a variable coefficient $\Delta\log R$ method by modifying the model coefficients and introducing new logging parameters to improve the applicability of the $\Delta\log R$ method to the evaluation of the organic quality of continental strata. Wang et al. (2016b) presented revisions along with application examples of the Devonian Duvernay shale in the Western Canada Sedimentary Basin to demonstrate the application of the improved $\Delta\log R$ method, which provided better and unbiased estimates of the TOC with a higher correlation coefficient for the shale gas evaluation. Zhao et al. (2017) proposed an improved model based on the original $\Delta\log R$ model and Wang's revision (Wang et al., 2016b), in which the approximate linear baseline is replaced by a theoretical one. Their results demonstrate that the improved model produces more reliable results in shale plays with large lithological variations. Maqsood et al. (2017) recommended using the average of the $\Delta\log R$ method using different log suits for the identification and quantification of the organic richness before sampling the intervals. However, the maturation history should be known before evaluating the organic matter richness of formation. Zhu et al. (2019) proposed the DD$\Delta\log R$ method in consideration of rock mineral composition variations and hole enlargement effects, and their prediction results were better than those obtained using machine learning algorithms. Wang et al. (2020) took the deep continental source rocks in the southwestern part of the Bozhong Sag as an example, and used a generalized $\Delta\log R$ method considering the density factor to predict the TOC content, which expanded the scope of the application of the traditional $\Delta\log R$ method. Tan et al. (2021) also reported that $\Delta\log R$ method is not suitable for TOC prediction of the shale reservoirs in the southern Sichuan Basin, and the application of multiple regression and natural gamma spectroscopy logging methods has the best effect in predicting the TOC content in this area.

3.4 Multivariate fitting method

Multiple logging parameters that are relatively highly correlated with the measured TOC content were performed to calculate the TOC, which are supposed to produce a more stable fitting linear equation. Based on the single parameter fitting results described in Section 3.2 and combined with the response characteristics of each logging curve to the organic matter, a variety of the TOC prediction schemes were designed using multiple linear regression methods. These schemes include binary linear fitting of 5 logging curve combinations, ternary linear fitting of 10 logging curve combinations, quaternary linear fitting of 10 logging curve combinations, five-element linear fitting of 5 logging curve combinations, and six-element linear fitting involving all of the highly TOC-correlated logging curves (Table 3). Since U spectral gamma logging has the best predictive effect in single factor analysis, the U curve was designed to participate in all of the fitting schemes.

Finally, the GR-U-DEN-CNL logging combination was optimized to perform quaternary linear fitting for the TOC estimation, because it had the best prediction results. The correlation coefficients (R) for the entire interval and for the objective interval in the Taiyuan Formation are as high as 0.78 and 0.97, respectively (Table 3). The fitted quaternary linear regression equation is shown in Eq. (11) and Eq. (12).

Entire interval:

$$\mathrm{TOC} = 0.01078\mathrm{GR} + 0.45691\mathrm{U} - 2.7344\mathrm{DEN} - 0.13181\mathrm{CNL} + 9.10822. \tag{11}$$

Objective interval:

$$\mathrm{TOC} = 0.00849\mathrm{GR} + 1.11637\mathrm{U} - 1.75443\mathrm{DEN} - 0.04988\mathrm{CNL} + 2.08154. \tag{12}$$

By observing the coefficients before each logging parameter, it was found that the changing features of TOC with each logging parameter are consistent with the previous single parameter fitting results, that is, the TOC is positively correlated with the values of GR and U, and is

Table 3 Pearson's correlation coefficient (R) between the TOC and multi-logging data for the studied samples from Well MY-1 in the entire interval and objective interval of Taiyuan Formation

Multivariate fitting	Entire interval		Objective interval	
	Logging combination	Correlation coefficient (R)	Logging combination	Correlation coefficient (R)
Binary	GR-U	0.57	GR-U	0.95
	AC-U	0.56	AC-U	0.94
	DEN-U	0.56	DEN-U	0.95
	CNL-U	0.61	CNL-U	0.92
	TH-U	0.58	TH-U	0.96
Ternary	GR-U-AC	0.58	GR-U-AC	0.96
	GR-U-DEN	0.58	GR-U-DEN	0.96
	GR-U-CNL	0.67	GR-U-CNL	0.96
	GR-U-TH	0.59	GR-U-TH	0.96
	DEN-U-CNL	0.73	DEN-U-CNL	0.96
	DEN-U-TH	0.59	DEN-U-TH	0.96
	CNL-U-TH	0.67	CNL-U-TH	0.96
	AC-U-DEN	0.56	AC-U-DEN	0.95
	AC-U-CNL	0.67	AC-U-CNL	0.94
	AC-U-TH	0.59	AC-U-TH	0.96
Quaternary	GR-U-AC-DEN	0.59	GR-U-AC-DEN	0.96
	GR-U-AC-CNL	0.68	GR-U-AC-CNL	0.96
	GR-U-AC-TH	0.59	GR-U-AC-TH	0.96
	GR-U-DEN-CNL	0.78	GR-U-DEN-CNL	0.97
	GR-U-DEN-TH	0.60	GR-U-DEN-TH	0.96
	GR-U-CNL-TH	0.67	GR-U-CNL-TH	0.96
	AC-U-DEN-TH	0.60	AC-U-DEN-TH	0.96
	AC-U-TH-CNL	0.69	AC-U-TH-CNL	0.96
	AC-U-DEN-CNL	0.76	AC-U-DEN-CNL	0.97
	DEN-U-CNL-TH	0.77	DEN-U-CNL-TH	0.97
Five-element	GR-U-AC-DEN-CNL	0.78	GR-U-AC-DEN-CNL	0.97
	GR-U-AC-CNL-TH	0.69	GR-U-AC-CNL-TH	0.96
	GR-U-AC-DEN-TH	0.61	GR-U-AC-DEN-TH	0.96
	GR-U-DEN-CNL-TH	0.78	GR-U-DEN-CNL-TH	0.97
	AC-U-TH-DEN-CNL	0.78	AC-U-TH-DEN-CNL	0.97
Six-element	GR-U-AC-DEN-CNL-TH	0.78	GR-U-AC-DEN-CNL-TH	0.97

negatively correlated with the DEN value. For the entire interval and for the objective interval, the TOC values calculated using the corresponding quaternary linear regression equations (TOC_MV) are in good agreement with the measured TOC results (Figs. 6 and 7). The predictive effect of the multivariate fitting method is significantly better than those of single parameter fitting method, and the $\Delta\log R$ method.

3.5 Comparative analysis of log-based TOC estimation

The reason why geophysical methods such as TOC prediction through logging data are feasible is that, in most cases, the increase in the organic matter content of a rock will directly affect the physical properties of the rock through the high resistivity, high gamma-ray, high acoustic transit time, and low density characteristics of the organic matter. Furthermore, TOC predications based on well logs are commonly utilized due to the continuous data availability for the entire zone of interest (Harris et al., 2019).

However, a difference inevitably exists between the TOC value predicted using the well logs and the values determined using laboratory geochemical analyses (Yu

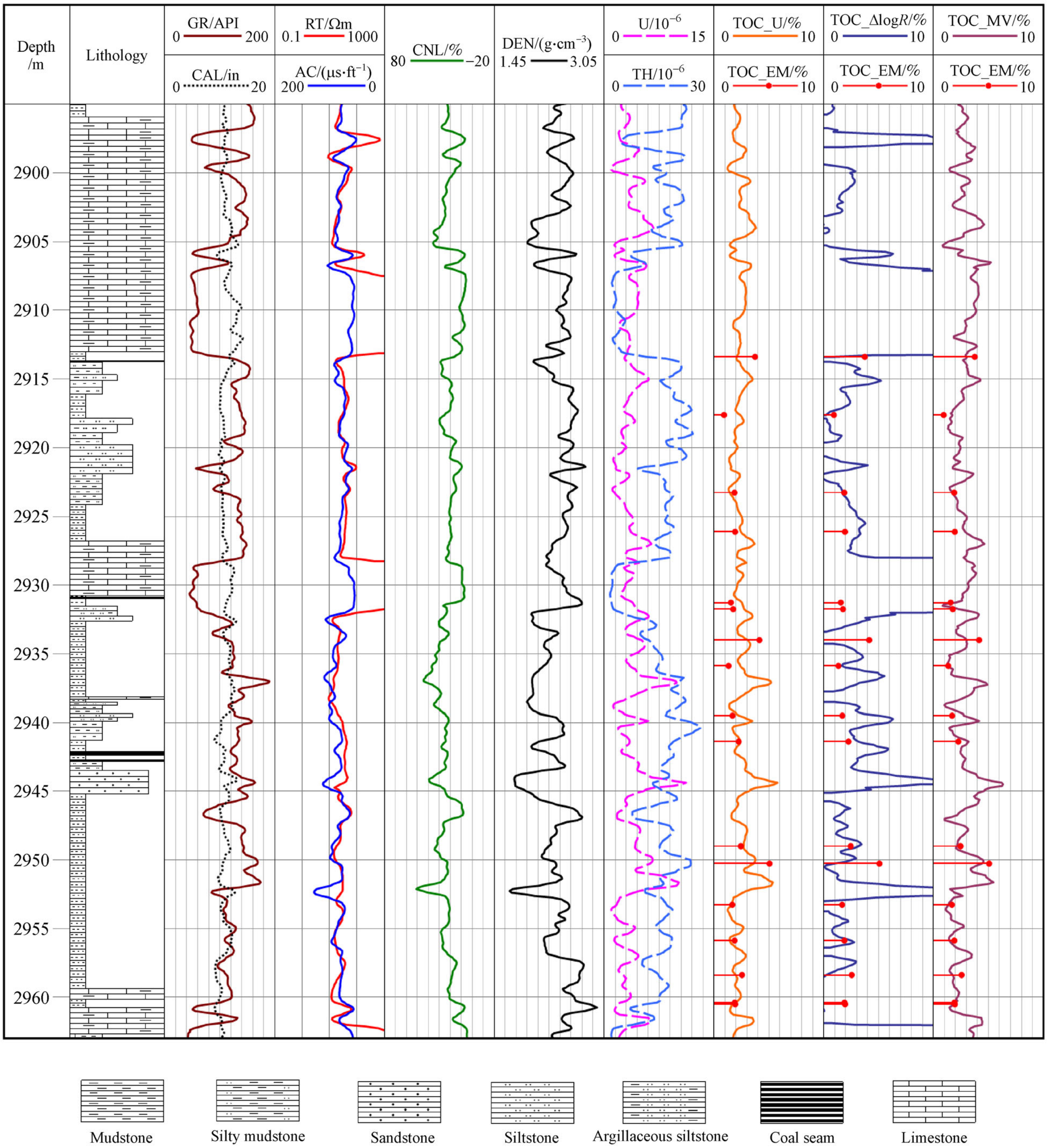

Fig. 6 Log data and comparison of the TOC prediction results obtained using different methods for the Taiyuan Formation in Well MY-1.

et al., 2017). Several well log-based methods are commonly used to predict the TOC, including single and composite well-log-based approaches. The latter includes the $\Delta\log R$ method (Passey et al., 1990) and the multivariate fitting method (Renchun et al., 2015). In this study, the single log-based technique prioritizes the U spectral gamma logging method, while the GR-U-DEN-CNL combination is preferred for the multivariate fitting method. The representative models of the three types of log-based TOC prediction methods were used to estimate the TOC values of the entire interval and of the objective interval in the Taiyuan Formation. The correlation between the experimentally measured TOC and the predicted TOC based on the well logs was investigated using evaluation indexes including the correlation coefficient (R), the coefficient of determination (R^2), the standard deviation,

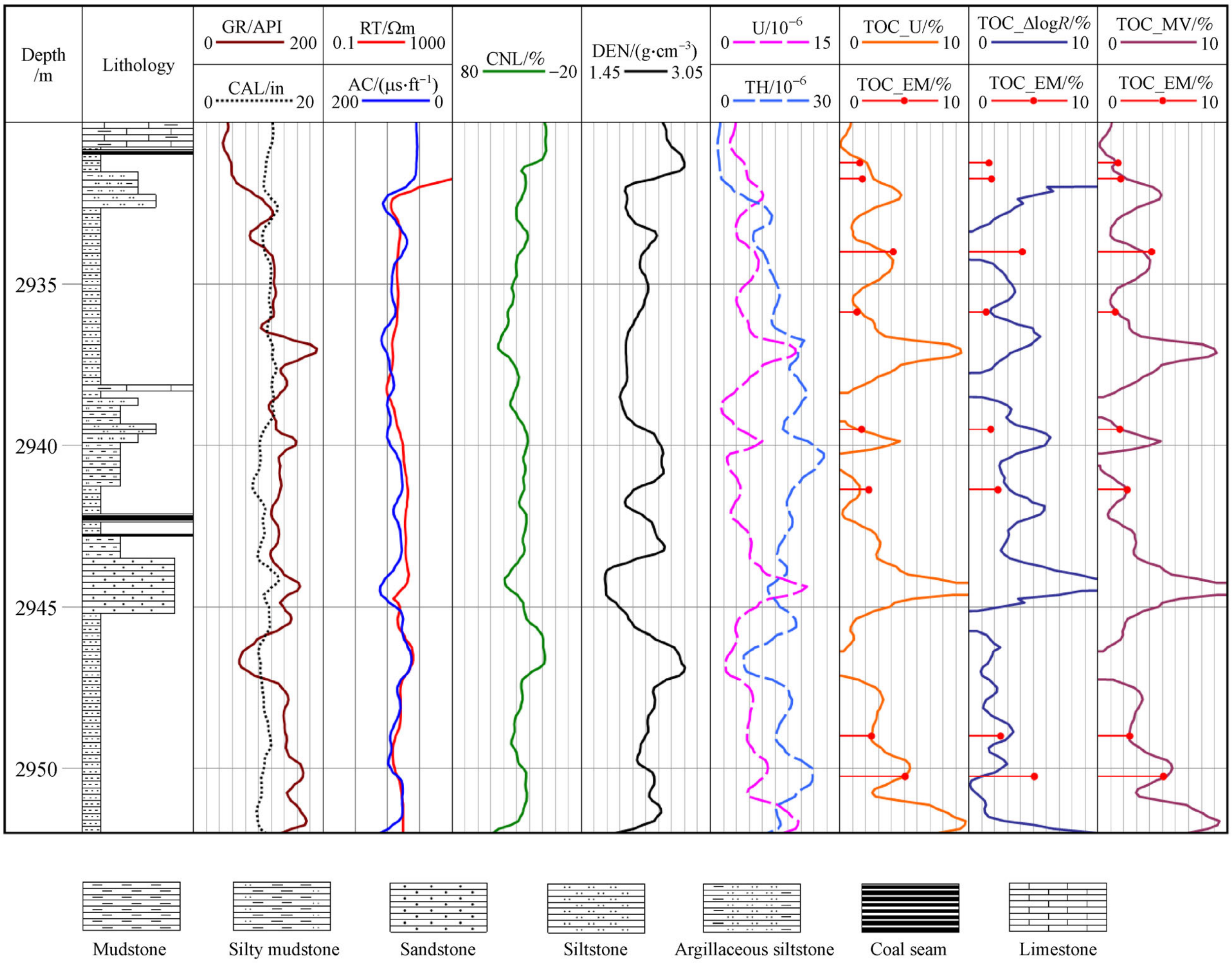

Fig. 7 Log data and comparison of the TOC prediction results obtained using different methods for the Taiyuan Formation in Well MY-1 at the target depths of 2930–2952 m.

the mean squared error (MSE) and the root mean squared error (RMSE) as shown in Table 4. From the shale sample analysis of the entire Taiyuan Formation, the average values of predicted TOC obtained using the U spectral gamma logging method, the Δlog*R* method, and the multivariate fitting method are (2.27±0.58)%, (3.1±6.05)%, and (2.27±0.81)%, respectively. The average TOC values predicted using the same approaches within the objective interval are (2.53±1.16)%, (6.25±7.4)%, and (2.53±1.23)%, respectively. The higher R and R^2 values and the lower standard deviation and RMSE values clearly demonstrate that the multivariate fitting of GR-U-DEN-CNL combination is the best-fitting method, and it predicts the TOC content of the marine-continental transitional shale in Taiyuan Formation with better accuracy (Table 4).

For the entire interval and for the objective interval, cross-plots were drawn of the measured TOC contents obtained using C/S analyzer versus the predicted TOC contents obtained using the U spectral gamma logging method, the Δlog*R* method, and the multivariate fitting method (Fig. 8). The R^2 values of the log-based TOC calculations of the different fitting methods and the measured TOC show that, the prediction effect of the multivariate fitting method is the best, followed by the U spectral gamma logging method. For the objective interval, the correlation coefficient of the spectral Gamma-ray logging method is 0.91, which is very close to the R value of the multivariate fitting method. Although the MSE and RMSE values of the U spectral gamma logging method are slightly higher than those of the multivariate fitting method, its standard deviation is lower than that of the multivariate fitting method, which shows that the single log-based technique of U spectral gamma logging method is also a better choice for predicting the TOC of the objective interval.

It was found from the above research that, when predicting the TOC for a high-quality reservoir similar to

Table 4 Linear regression statistics for the prediction of the TOC within the Taiyuan Formation in the study area

Taiyuan Formation	Regression statistics	U spectral gamma logging method	Δlog$R$ method	Multivariate fitting method
Entire interval	Calculated TOC	$\frac{1.45\text{—}3.62}{2.27}$*	$\frac{-3.19\text{—}21.52}{3.10}$*	$\frac{1.26\text{—}4.40}{2.27}$*
	Correlation coefficient (R)	0.55	−0.24	0.78
	Coefficient of determination (R^2)	0.31	0.06	0.61
	Standard deviation/%	0.58	6.05	0.81
	Mean squared error/%	0.75	41.42	0.43
	Root mean squared error/%	0.87	6.44	0.66
	Observations	17	17	17
Objective interval	Calculated TOC	$\frac{1.32\text{—}5.01}{2.53}$*	$\frac{-0.89\text{—}21.52}{6.25}$*	$\frac{1.38\text{—}5.31}{2.53}$*
	Correlation coefficient (R)	0.91	−0.53	0.97
	Coefficient of determination (R^2)	0.83	0.28	0.94
	Standard deviation/%	1.16	7.40	1.23
	Mean squared error/%	0.26	80.06	0.09
	Root mean squared error/%	0.51	8.95	0.30
	Observations	8	8	8

Note: *: $\frac{\text{Minimum—Maximum}}{\text{Average}}$.

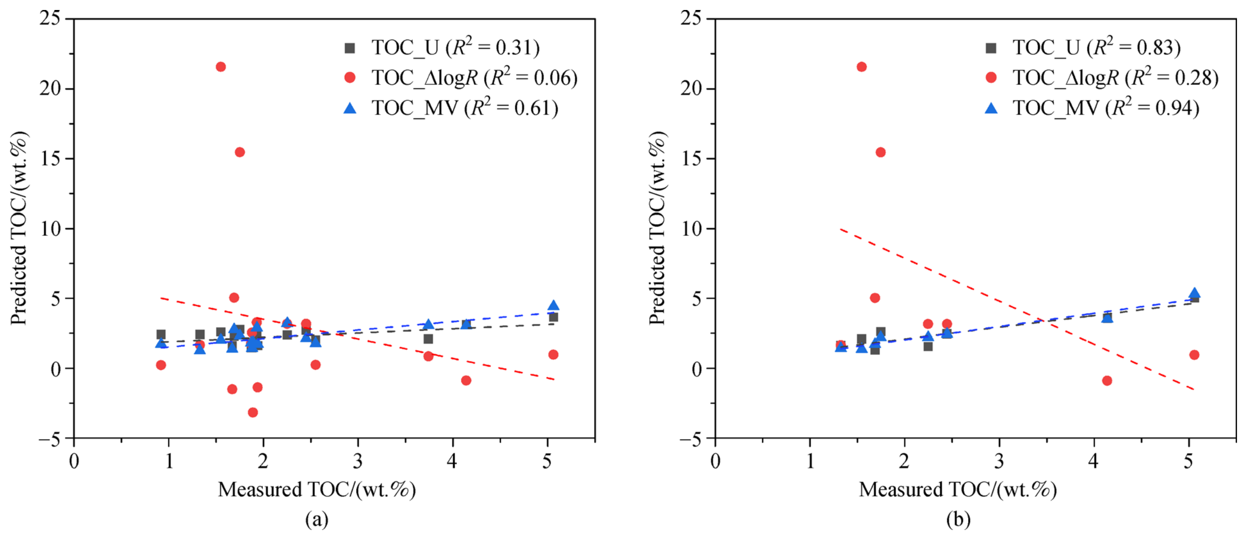

Fig. 8 Comparison of the predicted TOC values obtained using the well logging methods and the measured TOC for (a) the entire interval and (b) objective interval; the trend lines with different colors represent the corresponding best-fit line for each prediction method.

the objective interval, there is no need to perform multiple linear regression analysis with more than two parameters to obtain facilitate effective work, if a single parameter fits well, that is, the value of R^2 reaches 0.8 or more. For the transitional shale containing high-to-over-mature organic matter, the Δlog$R$ method is often not suitable for TOC prediction due to the constantly changing lithofacies and sedimentary environment. The TOC prediction method should be selected according to the actual situation of the study area.

4 Conclusions

The TOC content of the transitional shale in the Taiyuan Formation in the Zhongmu Block in the southern North China Basin was estimated using three types of well log-based methods, including the U spectral gamma logging, Δlog$R$, and multivariate fitting methods. Organic geochemical experiments were also performed on selected core samples from Well MY-1 for comparison with the logging prediction results and for the evaluation of the hydrocarbon generation potential. The following conclusions were obtained through a series of error analyses.

1) The organic matter in Zhongmu Block is highly mature to over-mature, corresponding to the period of dry gas generation. The vitrinite reflectance (R_o) of the selected shale samples ranges from 3.34% to 3.59%, with an average of 3.5%. In the study area, the organic matter has a strong hydrocarbon generation potential with kerogen type III, and the TOC content is relatively high.

2) The TOC content increases as the GR, AC, U, and TH values increase, and decreases as the DEN value increases, while the TOC and CNL do not exhibit a good positive correlations due to the digging effect in the highly gas-saturated layer. The single parameter fitting results show that U spectrum gamma logging has the best fitting effect for both the entire interval (R = 0.55) and the objective interval (R = 0.91). In addition, spectral gamma logging data are helpful for identifying the depositional environments of shale reservoirs, which has reference value for the comprehensive evaluation of shale gas.

3) The multivariate fitting method of the GR-U-DEN-CNL combination is considered as best due to its accurate prediction of the TOC, which is indicated by the highest values of correlation coefficients between the predicated and measured TOC values in the entire interval (R = 0.78) and the objective interval (R = 0.97), as well as the lower values of standard deviation, MSE and RMSE.

4) The TOC prediction methods applicable to different regions are also limited because of differences in the specific areas or geological formations. The effect of the multiple linear fitting is preferable in this study, and the most commonly used $\Delta\log R$ method is not applicable. It should be noted that, when predicting the TOC of high-quality reservoirs such as the objective interval investigated in this study, the single parameter fitting method with the value of R^2 more than 0.8 is nominated as a potential candidate for facilitating effective work.

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