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Gamma-ray spectrometry as a tool for stratigraphic correlations in the carbonate-dominated, organic-rich, pelagic Albian sediments in Central Italy

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Keywords: Albian, black shales, gamma-ray, spectrometry, stratigraphy, Umbria-Marche

ABSTRACT

Portable gamma-ray spectrometry provides a valuable method for detecting lithological variations and establishing correlations in monotonous sedimentary sequences. It has been applied to the pelagic, organic-rich and carbonate-dominated “Marne à Fucoïdi” Formation in the Marche-Umbria Basin. The relationship between the natural radioactivity parameters (potassium, thorium and uranium) and the carbonate and organic contents has been analysed in a reference section. The potassium and thorium signals appear as very good indicators of the clay content, whereas the uranium signal is not as straightforward. The latter is influenced by the content in organic matter, the thickness of black shales and the type of rocks alternating with black shales (marls or carbonates). In marl-dominated rocks, the uranium curve is a good indicator of the organic-rich zones, whereas, in carbonate-dominated sediments, the signal is more subtle and zones with a higher organic content do not always appear as good log markers. The total gamma-ray signal is strongly influenced by the potassium and amplifies the effects of the clay content with respect to the organic matter.

The validity of field gamma-ray investigations as a lithological and intra-basin correlation tool is confirmed by the excellent match between the total gamma-ray curve of the reference section, that from another field section and geophysical logs from a petroleum well. The precise correlation of field gamma-ray data with subsurface logs permits results from field studies (e.g., biostratigraphic boundaries) to be directly plotted onto the well data.

RESUME

Les spectromètres à rayons gamma portables permettent de détecter des variations lithologiques et d'établir des corrélations dans des séquences sédimentaires monotones. De telles mesures ont été effectuées dans les sédiments pélagiques, à prédominance carbonatée et riches en matière organique, de la Formation des “Marnes à Fucoïdes” dans le bassin de Marches-Ombrie. La relation entre les paramètres de la radioactivité naturelle (K, Th et U) et le contenu en carbonate et matière organique a été analysée dans une coupe de référence. Les réponses du potassium et thorium apparaissent comme de très bons indicateurs de la teneur en argiles. Le signal de l'uranium n'est pas aussi simple à interpréter, car il est influencé par le contenu en matière organique, l'épaisseur des black shales et la nature des sédiments alternant avec ces derniers. Dans les sédiments à prédominance marneuse, la courbe de l'uranium est un bon indicateur des zones riches en matière organique, alors que, dans les sédiments à prédominance carbonatée, le signal est plus subtil et les intervalles à plus forte teneur en matière organique ne sont pas toujours clairement exprimés. Le signal du rayonnement gamma total est fortement influencé par le potassium et amplifie les effets du contenu argileux par rapport à ceux de la matière organique.

Les études de terrain utilisant les rayons gamma sont un excellent outil de corrélation lithologique à l'intérieur d'un bassin sédimentaire, comme le démontre l'excellente correspondance entre les courbes du rayonnement gamma total dans deux coupes de terrain et les diagraphies géophysiques d'un puits pétrolier dans le bassin de Marches-Ombrie. La corrélation précise entre les données de rayons gamma acquises sur le terrain et les diagraphies de subsurface permet de placer directement sur les données de forage les résultats d'études de terrain (par exemple des limites biostratigraphiques).

Introduction

Gamma-ray logs are a measure of the natural radioactivity of rocks, which originates mainly from potassium (K), thorium (Th) and uranium (U). Such logs have been used extensively for decades by petroleum geologists to interpret lithology and correlate wells in the subsurface. More recently, the use of portable gamma-ray spectrometers has demonstrated the potential of this method in surface geology. This technique can contribute to stratigraphic correlations between outcrop sec-

tions or between outcrops and boreholes (van Buchem et al. 1992, Parkinson 1996, Bessa & Hesselbo 1997, Ahmadi & Coe 1998), highlight sedimentary cyclicity (van Buchem et al. 1992, Davies & Elliott 1996, ten Veen & Postma 1996,) or help unveil the origin of some sedimentary rocks (Myers 1989). The occurrence of radioelements relative to the organic carbon content has also been addressed in marine mud rocks (Myers & Wignall 1987).

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Fig. 1. Umbria-Marche Basin, Central Italy: location map of the Contessa (geographical coordinates: $x = 12^{\circ} 33' 56''$ E, $y = 43^{\circ} 22' 46''$ N) and Monte Petrano (geographical coordinates: $x = 12^{\circ} 36' 47''$ E, $y = 43^{\circ} 30' 10''$ N) field sections and of the Monte Cassiano well (geographical coordinates: $x = 13^{\circ} 23' 20''$ E, $y = 43^{\circ} 22' 35''$ N).

Radioactive elements tend to occur as major components of detrital minerals, whereas pure carbonates contain negligible amounts of K, Th and U (Adams & Weaver 1956, Mc Roberts et al. 1997). Consequently, most of the previous studies concentrated on siliciclastic sediments. This paper presents a case study in carbonate-dominated rocks, the organic-rich, pelagic sediments of the Albian part of the "Marne a Fucoidi" Formation deposited in the Umbria-Marche Basin of Central Italy (Fig. 1).

This study aims at analyzing the relationships between the natural radioactivity parameters (K, Th, U) and the carbonate and organic contents in the reference section of Monte Petrano (Fig. 1). By comparing the total gamma-ray signal of this section with those obtained from another field section and a petroleum well in the same basin, the potential for lithostratigraphic correlation will be demonstrated. Subsequently, the biostratigraphic boundaries precisely established in the field can be directly plotted onto the well log.

Geological setting

The Albian deposits of the Umbria-Marche Basin are characterized by dm-thick alternations of hemipelagic marls and limestones containing numerous, a few cm-thick intercalations of black shales (Arthur & Premoli Silva 1982, Herbert & Fischer 1986, Pratt & King 1986, Tornaghi et al. 1989, Fiet 1998). Black shales are defined herein as black, marly, laminated or bioturbated intervals, which have a content in total organic matter (TOC) higher than that of the encasing rocks. The interval studied comprises a lower marl-dominated and an upper carbonate-dominated unit (Fig. 2). This sedimentary sequence can be observed in many, stratigraphically continuous and

well-correlatable outcrops (Fiet 1998) and is well-dated by planktonic foraminifera (Coccioni et al. 1989, Tornaghi et al. 1989, Premoli Silva & Sliter 1994). Moreover, this sequence can be correlated with subsurface data from a nearby well (Fig. 1).

TOC values in black shales vary between 0.3 and 6 wt. % in the studied interval. Organic matter is mainly of continental origin (Pratt & King 1986, Fiet 1998). The clay mineral assemblages are homogeneous throughout the sequence: ca. 50% smectite, 30% illite and 20% mixed layers illite-smectite. Chlorite and kaolinite are absent and no glauconite-rich interval is present (Coccioni et al. 1989).

A detailed study (Fiet 1998) has shown the possibility for bed-to-bed lithological correlations with a less than half a metre resolution between field sections up to 40 km apart. The same applies to the biostratigraphic boundaries based on planktonic foraminifera. The most continuous field section at Monte Petrano (Fig. 1) has been used as reference section in this study (Fiet 1998). It is located some 450 m south-east of the Roccaccia culmination (geographical coordinates: $12^{\circ} 36' 47''$ E, $43^{\circ} 30' 10''$ N, sheet no. 116 IV SE, Cagli, topographical map of Italy, 1:25'000). It has been correlated with that of the Contessa quarry, some 20 km south (geographical coordinates: $12^{\circ} 33' 56''$ E, $43^{\circ} 22' 46''$ N, sheet no. 116, Gubbio, geological map of Italy, 1:100'000), and with the Monte Cassiano well, 50 km to the ESE (Fig. 1, geographical coordinates: $13^{\circ} 23' 20''$ E, $43^{\circ} 22' 35''$ N).

In the Monte Petrano section, the studied interval is about 51 m thick (Fig. 2) and spans the Early-Late Albian time interval (Coccioni et al. 1989). The lower marly E1 unit is about 25 m thick and consists essentially of alternating marls and black shales. The average sediment accumulation rate during the Albian is estimated at 5.9 m/myr (Fiet 1998). The upper lithological unit E2 is made of alternating marls and limestones (or marly limestones). Marls commonly grade to black shales.

Methods

TOC and CaCO_3 measurements

The TOC content (in wt. %) of each of the ca. 150 black shales identified in the Monte Petrano section was measured using a Leco device. This instrument determines the total carbon content of a rock, from which the carbonate carbon content (obtained with a Bernard calcimeter) was deducted, in order to obtain the TOC content. In this calculation, we assume that dolomite is not present in the carbonate fraction.

Natural radioactivity measurements (U, Th, K)

Natural radioactivity was measured in outcrops using a 4-channel portable GR spectrometer (Exploranium GR-256), which uses a GPS-21 detector thallium crystal. The spectrometer displays K, U and Th in counts per time period and converts these results to % K, ppm U and ppm Th. The detector is

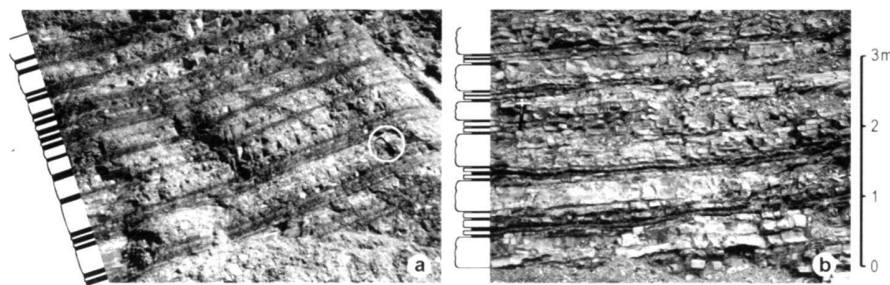


Fig. 2. The two main types of lithologies in the Albian outcrops of the Contessa quarry: (a) marl-dominated interval, (b) limestone-dominated interval. The alternations black shales/marls or black shales/limestones are well expressed in both lithologies.

placed in contact with a planar rock surface and remains stationary whilst measuring. The volume of rock sampled corresponds to a half sphere of 25 cm radius.

Natural radioactivity was measured every 35 cm. In limestones, fresh surfaces were used, whereas marls were dug in some 20 cm deep in order to avoid surface weathering. The sampling interval permitted the acquisition of a fairly continuous signal, reflecting lithological alternations a few decimetres thick. After numerous tests with count times up to five minutes, count times of two minutes were used, allowing for stable and reproducible measurements. The range of uncertainty in the measurements was estimated by carrying out sets of 20 repetitive measurements in the three types of lithology (limestone, marl, black shale): it reaches $\pm 0.1\%$ for K, ± 0.3 ppm for U and ± 0.5 ppm for Th.

K and Th are essentially contained in the detrital fraction of sedimentary rocks (Myers & Wignall 1987, Myers 1989, van Buchem et al. 1992). In carbonate-dominated environments they may be used to characterize variations in clay content: this is particularly true for the pelagic sediments studied where the contribution of radioactive heavy minerals (Hurst 1990) can be considered as insignificant. Uranium, which may be brought in with the detrital fraction, can also be precipitated from solution. In particular, it can be absorbed by organic matter and may be used to detect intervals enriched in organic matter (Myers & Wignall 1987, ten Veen & Postma 1996).

Results

Natural radioactivity in the Monte Petrano section (Fig. 2)

Potassium and thorium

The potassium content at this site varies between 0.2 and 3% (Fig. 3). Its stratigraphic variation is the mirror image of that of the carbonate content. The highest values are located in the basal, marly interval, where clay minerals are dominated by interstratified illite-smectite (Coccioni et al. 1989). Above this basal section, the potassium content progressively decreases towards the top of the section. This decrease coincides with the increase in carbonate content. In the marly unit E1, the clay content is over 50%, even reaching 100% in some levels. In the carbonate-dominated unit E2, the carbonate content is over 60% and the potassium content below 1%.

The thorium content varies between 2 and 7.5 ppm. Its stratigraphic variation resembles that of the potassium: a progressive decrease towards the top of the section, mirroring the progressive increase in carbonate content. The K/Th ratio is particularly stable and averages 0.25 throughout the section. This uniformity confirms the similar trend of both parameters.

Uranium

In the Umbria-Marche Basin, Albian black shales are distributed throughout the succession and arranged in bundles. Each bundle contains an average of three black shale levels (Fiet 1998), each a few centimetres thick. Within each bundle, black shales are separated by green marly intercalations or marly limestones, each a few centimetres thick (Fig. 2). The interval separating two bundles of black shales is always equal, or greater in thickness than the underlying black shale bundle. Therefore, the wavelength of this black shale/marl (or/limestone) rhythmic sedimentation averages 35–45 cm (Fiet 1998) and the sampling interval of gamma-ray measurements is in tune with this rhythmicity. This allows lithological variations to be analyzed at different scales and the response of the uranium signal to be tested without interferences with respect to the TOC content and total gamma-ray signal.

The uranium curve displays two different responses in the E1 and E2 lithological units. In the marly E1 interval, the average uranium content is about 1 ppm, with numerous peaks showing higher values. In the more calcareous E2 interval, maximum values barely reach 1 ppm. This reflects the significant lithological differences between the E1 and E2 units: the proportion of black shales is twice as high in E1 (ca. 100 levels over 25 m) as in E2 (ca. 50 levels over 25 m). Moreover, the lithology of the rocks alternating with the black shales differs: in unit E1, it is dominated by marls and, in unit E2, by marly limestones and limestones, which are usually poor in uranium (Myers & Wignall 1987). The combination of these two factors strongly decreases the uranium signal in unit E2. In the interval 36–40 m, a slightly stronger uranium signal coincides with both a higher concentration of black shales and a slightly more argillaceous section.

In the E1 interval, six major peaks in TOC have been labelled P1 to P6. These black shale horizons have TOC values over 2%. Only three of these peaks (P1, P3 and P5, containing

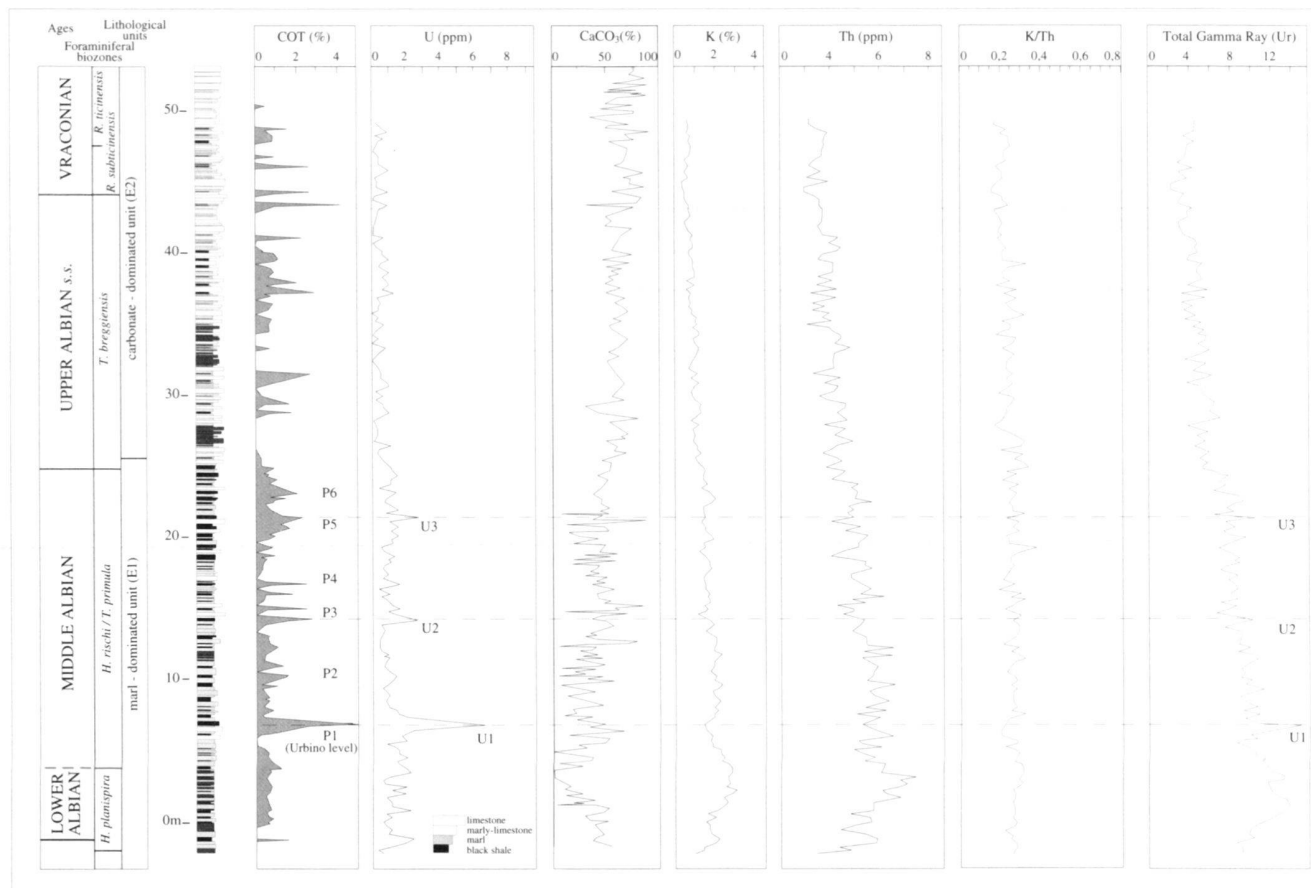


Fig. 3. Monte Petrano field section: lithology, biostratigraphy, geochemistry (organic and carbonate contents) and natural radioactivity (U, K, Th, total gamma-ray).

P1 to P6 correspond to major TOC (= total organic carbon) peaks in the marl-dominated E1 unit, and U1, U2 and U3 to significant uranium peaks in the same lithological unit.

respectively 5, 3 and 2.8 % TOC) match with significant uranium peaks, labelled U1, U2 and U3 (containing respectively 7, 3 and 3 ppm U). Therefore, organic-rich horizons are not always associated with uranium peaks. This difference originates in the way uranium and TOC measurements are acquired: whereas TOC values are limited to a very small rock sample, the portable gamma-ray spectrometer averages radioactivity over a rock volume corresponding to a half sphere of 25 cm radius. The uranium reading will be related to the TOC variations over the rock volume tested by the spectrometer. This relation is not linear, because the spectrometer emphasizes radioactivity readings at the centre of the sphere of investigation.

The U1 peak is associated with the Urbino level (TOC peak P1), a ca. 34 cm thick black shale interval of regional extension (Coccioni et al. 1987), which has an average TOC content over 5%. All other black shales are less than 10 cm thick in the studied interval. The U2 and U3 peaks correspond to "elementary bundles" (*sensu* Fiet 1998) and contain an average TOC content of over 2%.

Consequently, the uranium signal in the carbonate environment of the Marche-Umbria Basin is regulated by the presence and thickness of black shales, whose TOC content may vary considerably, but also by the nature of the encasing rocks (marls or limestones) and the sampling rate. These observations demonstrate that the use of uranium logs is not as straightforward as that of the potassium and thorium.

In a marly environment (average carbonate content of less than 50%), the uranium signal is a good indicator of the main variations in TOC content: in the basin, significant increases in uranium correspond to major levels of organic matter accumulation. In a carbonate-dominated environment, variations in the uranium content are more subtle and difficult to use as a lithological indicator.

Total gamma-ray

The relative contribution of potassium, uranium and thorium to the total radioactivity is calibrated for the "ur", or unit of ra-

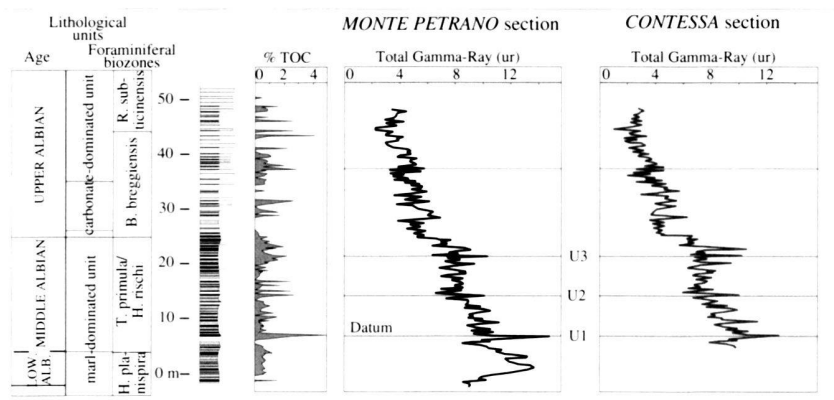


Fig. 4. Correlation of outcrop gamma-ray data from the two field sections at Monte Petrano and Contessa.

U1, U2 and U3 correspond to significant uranium peaks in the the marl-dominated E1 unit (see Figure 3).

dioelement concentration (International Atomic Energy Agency 1976). One ppm U is equivalent to 1 ur, 1% K is equivalent to 2.6 ur, and 1ppm Th is equivalent to 0.477 ur (van Buchem et al. 1992). Considering the uncertainties associated with the measurements carried out with the field spectrometer (see above), the range of uncertainty for the total gamma-ray can be estimated at a maximum of ± 0.7 ur.

Because of this calibration with the ur, the potassium content will significantly influence the total gamma-ray curve and, together with the thorium, amplify the effects of the clay content with respect to that of the organic matter. The general shape of the total gamma-ray curve (Fig. 2) resembles that of the thorium. The potassium curve is smoother than the thorium and total gamma-ray, but displays the same general trend. The E1 and E2 units are clearly distinct on the total gamma-ray curve. The E1 unit can be subdivided into three parts with a decreasing average total gamma-ray (12, 10 and 8 ur), related to the average increase in carbonate content. Each of these parts is bounded by the uranium peaks U1, U2 and U3.

Discussion

In order to validate the field gamma-ray data acquired in the Monte Petrano section, measurements have been compared with data obtained in another field section and with gamma-ray and sonic logs recorded in a petroleum borehole.

The Contessa field section is located in a quarry, some 20 km south of the Monte Petrano section (Fig. 1). Based on a detailed field study using cyclostratigraphy, these two sections can be correlated with a lithological resolution of less than 10 cm (Fiet 1998). No sedimentary hiatus has been observed over the studied interval in the Contessa section. Gamma-ray measurements were carried out in a similar way to those in Monte Petrano.

The comparison of the total gamma-ray signal in both sections shows a high degree of similarity (Fig. 4). In both sections, a sharp break marks the boundary between the E1 and

E2 lithological units, and the U1 to U3 peaks can be clearly identified. The general trend of the curve, as well as the distinctive peaks seem to be even better expressed in the Contessa section. This is probably related to the activity of the quarry, where fresh outcrops are regularly exposed. The significant horizons highlighted in the Monte Petrano section, in particular those rich in organic matter, display the same gamma-ray signature as in the Contessa section and provide valuable markers for intra-basin correlations.

The total gamma-ray signature in Monte Petrano has been subsequently compared with the gamma-ray and sonic logs of the petroleum well Monte Cassiano (Fig. 5) located in the same basin, some 50 km to the east-southeast (Fig. 1). This well has been chosen because it penetrated a thick "Marne a Fucoidi" section. The sonic log emphasizes the different physical properties of limestones, marls and black shales.

No sedimentary hiatus or syndimentary faults have been observed in this borehole. The datum used in the correlation with the Monte Petrano section is the major U1 peak identified at both locations and corresponding to the Urbino level, a regional marker. The trend on both well logs is similar to that obtained with the field spectrometer in Monte Petrano. The U1 to U3 peaks are clearly identifiable, as well as other peaks. There seem to be only minor thickness variations between the two locations. The gamma-ray provides a good correlation tool, even in the carbonate-dominated E2 unit. The use of the sonic log for correlation purposes is noteworthy in this carbonate interval.

This excellent match with well data emphasizes the contribution of field gamma-ray to stratigraphic correlations. It allows the biostratigraphic boundaries precisely established in the field using planktonic foraminifera (Fig. 5) to be directly plotted onto the well logs with a precision of less than one meter. On the other hand, the precise correlation with the stratigraphically continuous reference section of Monte Petrano confirms the absence of major sedimentary hiatuses in the well.

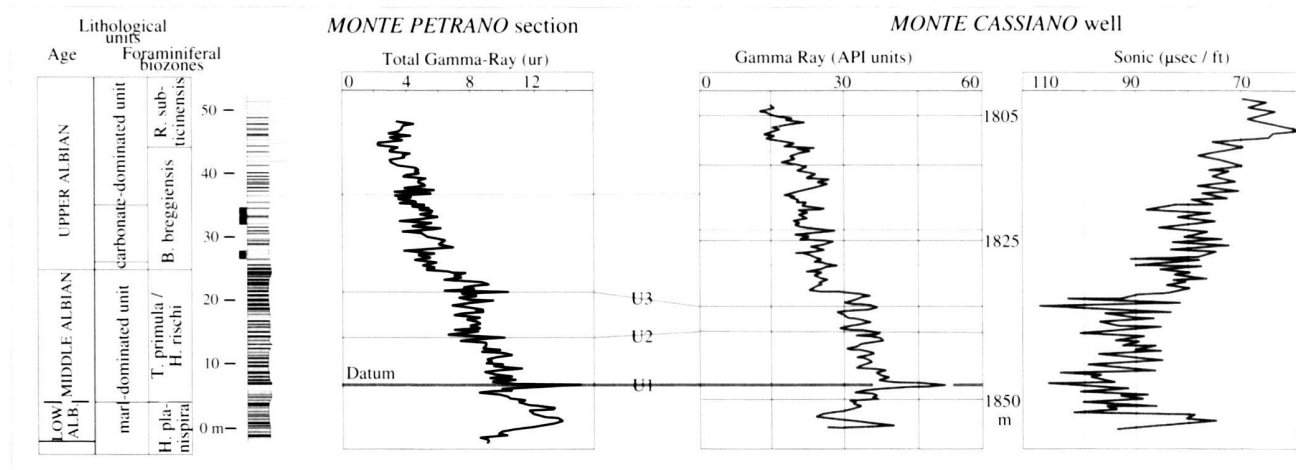


Fig. 5. Correlation of outcrop gamma-ray data from the Monte Petrano section with gamma-ray and sonic logs from the Monte Cassiano well. U1, U2 and U3 correspond to significant uranium peaks in the the marl-dominated E1 unit at Monte Petrano (see Figure 3).

Conclusions

The natural radioactivity of biostratigraphically and cyclostratigraphically well-calibrated pelagic sediments has been measured in the Marche-Umbria Basin using a portable gamma-ray spectrometer. The lithologies studied grade from marls to limestones, with intercalations of black shales.

In the first stage, the behaviour of the potassium, thorium and uranium signals was compared with the carbonate and organic contents in the reference section of Monte Petrano. Potassium and thorium are good indicators of the clay content and can be used as lithological indicators. The uranium signal depends upon different factors: the proportion of organic matter, the thickness of black shales and the type of sediments intercalated with black shales (i.e. marls or limestones). In marl-dominated sediments, the uranium signal is a good indicator of the major organic-rich intervals. In a carbonate-dominated environment, the use of uranium data is more subtle and horizons enriched in organic matter are not always highlighted.

In the second stage, the behaviour of the total gamma-ray curve was studied. The contribution of the three radioactive elements according to the international "ur" unit gives more weight to the potassium content and, consequently, the total gamma-ray amplifies the effects of the clay content. Nevertheless, in marl-dominated intervals, the major peaks detected in the uranium data are still visible, making it possible to identify organic-rich intervals on the total gamma-ray.

Finally, the validity of field gamma-ray logs in regional studies was demonstrated by comparing data from the reference section with those from another field section (Contessa section) and with logs recorded in a petroleum well (Monte Cassiano). The total gamma-ray curves are highly similar at the three locations, thereby enabling detailed intra-basin stratigraphic correlations. The major gamma-ray peaks can be used

as regional markers. Observations obtained from detailed field studies, in particular biostratigraphic boundaries, can be accurately plotted onto the well logs.

This case history demonstrates the contribution of field gamma-ray spectrometry to intra-basin correlations. Natural radioactivity parameters help detecting lithological variations and zones of major organic matter accumulation. Moreover, it provides a simple tool for correlating outcrop sections with subsurface data, which makes it particularly useful in petroleum exploration.

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