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Marlise Colling Cassel

IMPLICAÇÕES PALEOAMBIENTAIS DOS FOLHELHOS NEGROS NA RAMPA CARBONÁTICA DA FORMAÇÃO IRATI

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Marlise Colling Cassel

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Orientador: Prof. Dr. Ernesto Luiz Correa Lavina

Banca Avaliadora: Dr. Laury Medeiros de Araújo

Dr. Francisco M. W. Tognoli

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SUMÁRIO

APRESENTAÇÃO DA DISSERTAÇÃO

O estudo aqui apresentado é fruto do trabalho de mestrado executado na Universidade do Vale do Rio dos Sinos ao longo de dois anos, sob orientação do Professor Dr. Ernesto Lavina. A dissertação intitulada IMPLICAÇÕES PALEOAMBIENTAIS DOS FOLHELHOS NEGROS NA RAMPA CARBONÁTICA DA FORMAÇÃO IRATI tem seus resultados apresentados na forma de artigo científico intitulado: PERMIAN CARBONATE RAMPS AND BLACK SHALES: PALEOENVIRONMENTAL DYNAMICS IN A RESTRICT BASIN. O artigo apresenta um detalhamento das fácies e arcabouço estratigráfico da Formação Irati no sul da Bacia do Paraná, bem como inferências paleodeposicionais baseadas em análises de geoquímica orgânica e gamaespectrometria. Este artigo foi submetido ao periódico Palaeogeography, Palaeoclimatology, Palaeoecology, o qual pertence ao estrato A2 qualis CAPES. O trabalho tem a coautoria do orientador Dr. Ernesto L. C. Lavina e coorientadora Dra. Joice Cagliari, da Unisinos, e Dr. René Rodrigeus e Dr. Egberto Pereira, da Universidade do Estado do Rio de Janeiro, com os quais desenvolveu uma parceria de trabalho.

Introdução

Estudos envolvendo a deposição de folhelhos pretos remetem, de forma recorrente, uma associação com eventos de escala global no passado geológico. Mudanças climáticas, perturbações no ciclo do carbono, até extinções em massa podem estar bem registradas em intervalos de folhelhos negros, sobretudo quando depositados em rampas carbonáticas. Dessa forma, unidades estratigráficas que contemplem tais registros se mostram como uma ótima oportunidade de investigação de fenômenos extremamente relevantes do passado.

A Formação Irati, além de corresponder a estes pré-requisitos é um marco na Bacia do Paraná, dada sua extensa lateralidade faciológica. Cabe salientar que também é relevante para todo o interior cratônico do Gondwana, na medida em que é síncrona com outros pacotes de folhelhos ricos em matéria orgânica presentes nas bacias Chaco-Paraná, Karoo e Kalahari.

Sabendo da necessidade de mais estudos, discussões e detalhamento da Formação Irati, devido sua grande distribuição areal e restritas áreas aflorantes, o estudo aqui apresentado espera contribuir com o arcabouço estratigráfico e enriquecer discussões já existentes acerca da dinâmica paleoambiental do Gondwana no Permiano.

Objetivo

A partir da contextualização do objeto de estudo (a Formação Irati), destacam-se questões para problematização deste tema (sua dinâmica paleoambiental): a) é possível delimitar a influência do clima no seu empilhamento sedimentar? b) como se dá a variação de salinidade e oxigenação do substrato?

Desta forma foi delimitado como objetivo deste trabalho detalhar a evolução paleoambiental da Formação Irati, tendo a salinidade e oxigenação do substrato como possíveis indicadores do clima.

Métodos

Para execução do trabalho foi necessária a descrição de testemunhos de sondagem, e perfis geofísicos de raios gama para correlação, que resultou na confecção de perfis litológicos e uma seção estratigráfica, cujo datum é o nível inferior de folhelhos negros. Estes compuseram o arcabouço estratigráfico analisado na perspectiva das sequencias

deposicionais. A partir de então foram feitas coletas para análises de geoquímica orgânica e uma medição gamaespectrométrica, com o fim de diferenciar os canais de potássio, urânio e tório. No corpo do artigo submetido ocorre o detalhamentos das informações referentes aos materiais e métodos.

Resultados

Os resultados obtidos são apresentados na forma de artigo científico submetidos ao periódico Palaeogeography, Palaeoclimatology, Palaeoecology. Foi definido o sistema deposicional da Formação Irati como rampa carbonática com os domínios interno, médio e externo, como já proposto por Araújo (2001). O arcabouço estratigráfico foi detalhado em três sequencias deposicionais e ciclos T-R de mais alta frequência. Com relação aos aspectos paleoambientais foram traçadas inferências quanto à natureza e teor de matéria orgânica presente. Os dados de geoquímica orgânica, aliados aos dados de gamaespectrometria permitiram traçar inferências paleoambientais que evidenciaram oscilações no clima por meio da variação entre períodos de menor e maior umidade, bem como de concentração e diluição da salinidade. Cabe ainda destacar o registro fossílifero de escamas de peixe e restos esqueletais de mesossaurídeos.

Permian carbonate ramps and black shales: Paleoenvironmental dynamics in a restrict basin

Marlise Colling Cassel[1*], Ernesto Luiz Correa Lavina[1], Joice Cagliari[1], René Rodrigues[2], Egberto Pereira[2]

[1]*Universidade do Vale do Rio dos Sinos, Programa de Pós-Graduação em Geologia, Av. Unisinos, 950, Cristo Rei, São Leopoldo, Rio Grande do Sul, CEP 92022-000, Brazil*

[2]*Universidade do Estado do Rio de Janeiro, Departamento de Estratigrafia e Paleontologia, Rua São Francisco Xavier, 524, Maracanã, Rio de Janeiro, Rio de Janeiro, Brazil*

Abstract

Bituminous shales have been associated to worldwide events, such as mass extinction and anoxia, mainly when preserved in carbonate ramps, and constitute reliable records of this dynamic. The Irati Formation is very suitable for the study of these intricate processes due to the presence bituminous shales and carbonates. Based on core descriptions, gamma spectrometry data (K, U, Th) and organic geochemistry (TOC, R_{ins}, S, IH and IO) a detailed stratigraphic scheme was proposed for the southern sector of the basin. It is composed by three depositional sequences formed internally by T-R cycles of highest frequency. The analysis of the sedimentary facies demonstrated a carbonate ramp subdivided into outer, middle and inner. It was possible to define a climatic control of the sea level changes and climatic control, as well as oscillations in the salinity and oxygenation. The bituminous shales record regular salinity levels, with dysoxia levels and even euxinia, associated to the increase of the bioproductivity. The carbonate facies register periods of hypersalinity under oxic environment, under semi-arid conditions. It was evidenced that the accumulation of *Mesosaurus* skeletal remains results from the action of reworking fluxes, creating an endemic facies in this region.

Key words: High resolution stratigraphy. Permian. Intracratonic basins. *Mesosaurus*. TR sequences. Organic geochemistry. Gamma spectrometry. Anoxia

1. Introduction

Black shales have been associated to both regional and global geologic events, such as climatic changes, oceanic anoxia and mass extinction (Phelps et al., 2015). Anoxic events have been concomitant with significant perturbationn in the carbon global cycle, influencing both the atmospheric and oceanic CO_2 concentration (Bonis et al., 2010; Phelps et al., 2015). The most expressive records of these events are in carbonate platforms with occurrence of abrupt changes between carbonatic facies and anoxic shales (Phelps et al., 2015).

In the middle and upper Permian, black shales have been studied under the biogeochemical standpoint, because of the mass extinction at the end of this period (Wignall and Twitchett, 1996; Clapham and Payne, 2011). Some models propose a physiologic crisis resulting from the: pCO_2 and temperature increase stresses, associated to the pO_2 reduction and carbonate saturation which caused acidification and environmental anoxia. Besides, salinity changes are commonly associated to pCO_2 changes (Knoll et al., 2007).

In the Permian of the Paraná Basin and adjacent gondwanic basins of Chaco-Paraná (Argentina), Karoo and Kalahari (Southern Africa) occurs thick packages of shales rich in organic matter, intercalated with carbonatic layers (Oelofsen and Araújo, 1983; Oelofsen and Araújo, 1987; Lavina, 1991; Hachiro, 1996; Milani et al., 1998Araújo, 2001). In the Permian there are black shales in several basins of the cratonic interior of the South America which respond to the same tecto-sedimentary cycle (Soares et al., 1974). This cycle of continental expression is synchronous with huge sedimentary units of the center-east of the North America, as proposed by Sloss (1963).

48 The Irati Formation is a suitable example to be studied as part of the worldwide
49 problematic aforementioned, since it has been deposited in the Middle Permian and is
50 composed by organic shales, siltstones and carbonates (Rocha-Campos et al., 2006; Santos et
51 al., 2006). Therefore, this formation can be a reliable register of the Permian environmental
52 dynamics which has been characterized by extreme events.

53 However, due to its broad area (around 2000 km in the N-S axis) and faciologic
54 variations associated to the outcropping area restricted shortly to the basin margin, the Irati
55 Formation still needs a further detailing in paleoenvironmental interpretations. Previous
56 studies have not reached agreement in paleoenvironmental aspects, being the salinity one of
57 the main topics of debate.

58 The main purpose of this work is the proposal a high-resolution stratigraphic scheme
59 for the bituminous shales and carbonates of the Irati Formation, and the analysis of its
60 paleoenvironmental meaning, based on geophysical and organic geochemistry approaches.
61 This analysis intends to bring a contribution to the understanding of the global paleoclimatic
62 dynamics due to the close relation between black shales and global events. Last but not least,
63 it will also contribute for the comprehension of both high frequency climatic and eustatic
64 controls.

65 2. Methodology

66 Twenty-three cores from the Brazilian Geological Survey (CPRM) were described and
67 analyzed in the southern part of Parana Basin (Fig. 3). The faciologic scheme obtained was
68 analyzed under the sequence stratigraphy standpoint, according to Hunt and Tucker's (1992)
69 model.

70 In two cores, samples for total organic carbon (TOC), total sulfur (S) and insoluble
71 residual have been collected each 60 cm core depth, totaling 214 samples, which have been

analyzed at *Laboratório de Estratigrafia Química e Geoquímica Orgânica da Faculdade de Geologia, Universidade Estadual do Rio de Janeiro*. The R_{ins} was obtained by hot acidification for elimination of carbonates. A LECO-COT502-308 equipment was used for both TOC and S analyses. Subsequently, 52 samples have been chosen for pirolysis-rock eval analyses, which supplied hydrogen index (HI) and oxygen index (OI) values.

Additionally, new measurements in a gamma spectrometer RS 124 Radiation Solutions, with a collect period of three minutes each 50 cm was conducted. This allowed the differentiation of the potassium (K), thorium (Th) and uranium (U) channels and the ellaboration of new profiles, whose respective curves have been normalized to improve the comparison of trends.

3. Regional Geology

The Paraná Basin is an intracratonic basin that spreads through the central-south Brazilian territory, Northern Argentina, Paraguay and Uruguay, covering an area of 1 500,000 km² (Fig. 1). The Phanerozoic record reaches up to 8 km thick at its depocenter, ranging units from the Late Ordovician to the Late Cretaceous (Fig. 2). The evolution of those units is linked to uplifts and subsidence of orogenetic phases of the Southern Gondwana. The Paraná Basin is subdivided into six supersequences: (i) Supersequence Rio Ivaí (Ordovician–Silurian), (ii) Supersequence Paraná (Devonian), (iii) Supersequence Gondwana I (Carboniferous–Lower Triassic), (iv) Supersequence Gondwana II (Triassic), (v) Supersequence Gondwana III (Jurassic–Cretaceous) and (vi) Supersequence Bauru (Cretaceous) (Zalan et al., 1990; Milani, 1997; Milani and Ramos, 1998; Cagliari et al., 2014) (Fig. 1 and 2).

The Supersequence Gondwana I (Fig. 2) has a transgressive-regressive trend and a gradual paleoenvironmental change, from glacially influenced at the bottom towards arid ones

in the top (Milani, 1997). The sedimentation begins with the glacial deposits of the Itararé Group (Fig. 2). In the top of this group there is glacial conditions associated to the marine transgression (Palermo and Rio Bonito formations) (Lavina and Lopes, 1987). The Rio Bonito formation is composed of estuarine and deltaic deposits in the base, and facies of barrier-beach with lagoon bodies and marshes associated (including thick coal layers). The progressive relative sea level rising resulted in shoreface deposits (upper part of the Rio Bonito Formation) and offshore of the Palermo Formation (Fig. 2). The adjacent unit (Irati Formation) is composed by siliciclastic deposits with intervals rich in organic matter, carbonate and evaporites (Lavina, 1992). The Irati Formation is overlain by the Serra Alta and Teresina formations which represent the establishment of continental environments in the basin. The top of this supersequence (Rio do Rasto and Pirambóia formations) represent non-marine sedimentation, with lacustrine, fluvial and aeolian facies indicating the establishment of arid conditions along the Mesozoic (Lavina, 1988; Lavina, 1992).

The Irati Formation, of Artinskian age (278.4 ± 2.2 Ma) (Santos et al., 2006) has fine siliciclastic sediments, occasionally rich in organic matter, intercalated with carbonates and evaporites. The unit is divided into two members: Taquaral (lower) and Assistência (upper). The first is composed by claystones and siltstones, while the second member has siltstones and black shales, ocasionally intercalated with carbonates (Mendes et al., 1966; Padula, 1968; Petri and Coimbra, 1982; Oelofsen and Araújo, 1983; Lavina, 1992; Hachiro, 1996; Milani et al., 1997; Araújo, 2001). In general terms, the sedimentation of the Irati Formation is considered as a marine restrict epicontinental environment, with salinity fluctuations, substrate oxygenation and bioproductivity (Lavina, 1992; Hachiro, 1996; Araújo, 2001; Rodrigues et al., 2010a; Rodrigues et al., 2010b). It is a depositional system of carbonate ramp, divided into external, middle and inner ramps (Araujo, 2001). This association

120 represents the sedimentation during the restriction of the marine influx in the Paraná Basin
121 (Lavina, 1992; Milani et al., 1997).

122 The black shales and carbonates of the Assistência Member have been extensively
123 analyzed. They present high values of total organic carbon (TOC) up to 22% in the shales,
124 and the intercalated carbonate with low values of insoluble residues (Padula, 1968; Araújo,
125 2001; Rodrigues et al., 2010a; Rodrigues et al., 2010b; Alferes et al., 2011). The Assistência
126 Member is also characterized by the occurrence of skeletal remain of *Mesosaurus*, which can
127 be correlated to the Whitehill Formation, in the South Africa (Oelofsen and Araújo, 1983;
128 Oelofsen and Araújo, 1987; Lavina et al., 1991; Lavina, 1992; Soares, 2003).

129 4. Results and interpretations

130 4.1 Depositional System

131 The facies succession in the core description demonstrated the occurrence of a
132 carbonate ramp, as characterized by Burchette and Wright (1992), Wright and Burchette
133 (1996), Bosence and Wilson (2003), Miall (2010). This ramp is divided into outer, middle and
134 inner domains, all recorded in the deposits herein studied.

135 In the outer ramp (OR) domain, located below the level of the storm waves action,
136 predominates fine siliciclastic sediments, where may occur dysoxia. For the studied deposits
137 the following facies have been described: OR1) Black shale rich in organic matter (Fig. 4A).
138 Presents high peaks of TOC (around 20%), very high total S (1 to 3.5%) and R_{ins} around 75%
139 (Fig. 6); OR2) argillaceous siltstone with deep gray fissility, sometimes medium with
140 incipient lamination (Fig. 4B). It has rare fossiliferous levels with fish scales and crustaceans
141 in its upper part (Fig. 4F). The values of TOC are lower than 1%, the S ones are lower, and
142 the R_{ins} is high (85%) (Fig. 6); OR3) Siltstone gray with sparse wavy bedding planes of very
143 fine sandstone (Fig. 4C). It presents both TOC and S values very low, and R_{ins} values very

high (Fig. 6); OR4) Heterolithic claystone and very fine sandstone. Interbedding of gray claystone with milimetric to centimetric lenses of fine sandstone occur (0.1 to 2 cm) (Fig. 4D). The claystone-sandstone contact is abrupt, being the sandstone and the claystone layers truncated. The sandstone-claystone contacts, on the other hand, are either abrupt or gradational. It has low TOC values, occasionally medium to high, with oscillatory pattern of S and high values of R_{ins} (Fig. 6). In specific levels there are skeletal remains of *Mesosaurus* (Fig. 4G). It is noteworthy that in 22 out the 23 cores described ribs, vertebrae, limb bones and teeth of mesosaurids were found (Cassel and Lavina, 2013).

The physical processes involved in this domain are characterized by the predominance of decantation. It has been identified, moreover, the influence of distal oscillatory fluxes and reworking currents (bottom of storms), evidenced in the OR4 facies due to the disarticulation of the bone fragments (Cassel and Lavina, 2013).

The middle ramp (MR) is composed by both carbonate and siliciclastic sedimentation, whose alternance is typical of the study area. Occurs trough the gradation from claystone to carbonate, corresponding to shallowing pulses (Fig. 4E). Other shallowing evidences have also been found: tepee (Fig. 4H) and salt layer (Fig. 4I), similar to anhydrite nodules. Over this interval there are carbonate with low angle, usually with wavy lamination on the top (wave ripples and hummocky crossed stratification). The intercalated siliciclastic intervals are composed of argillites, occasionally with layers of very fine sand and shale. Usually, present low TOC with peaks oscillating toward higher values, but not exceeding 2%. The S is variable but predominantly low, while the R_{ins} has a saw-pattern between 15% and 85% (Fig. 6).

The physical processes in this domain demonstrate the action of oscillatory fluxes, reworking currents (either bottom or storms). In relation to the shallowing evidences found, mainly the most intense (tepee), due to its low thickness and occasional occurrence, has also

been placed in the middle ramp. Being a ramp with smooth declivity, ephemeral relative sea level oscillations may cause exposition even in this domain, as well as drownings.

The inner ramp (IR) is a shallow water environment of where carbonatic sedimentation takes place, subjected to subaerial exposition. In the study area, it were described carbonatic breccias with centimetric angulose clasts (Fig. 4E). The clasts of this breccias present lamination in domic microrelief characteristic of microbialites (Araújo, 2001). The breccias and the V-shapped molds are evidences of subsequent aerial exposition. They present low values of TOC and R_{ins}, and high values of S (Fig. 6).

4.2 Organic matter, oxygenation and salinity (organic geochemistry and gamma spectrometry)

The TOC values of the Irati Formation are characterized by two aspects: Present levels rich in organic matter (> 1%) (Arthur and Sageman, 1994) distributed along all the succession and two anomalous occurrences of high peaks (around 20%) (Fig. 6).

The kerogen was characterized as type II, and subordinately as type III, through the IH and IO data (Tissot and Welte, 1984) (Fig. 5). It was not recorded any abundance of organic matter rich in lipidic fractions with H (the most reactive fraction) to reach Type I levels. This means that the organic matter herein studied is either variably degraded, or of marine origin (Type II) and subordinately continental (Type III), in a mixtured or alternated pattern.

The IH/TOC graphic demonstrates an asymptotic pattern, where the increasing input of organic matter is deprived of the corresponding increase in IH (Fig. 5). From this asymptotic point onwards (IH=300 and COT=4), whose values belong to the bituminous shale facies (OR1) (Fig. 4A), variations of the precursor source may occur, or delimitate the beginning of the selective biodegradation. Palynological data indicate that from this TOC level onwards (3% to 20%) occurs the highest amount of amorphous organic matter of the

193 interval, with significant content of H (Araújo, 2001). This demonstrates that the analyzed
194 kerogen comes from H-rich sources partially biodegraded, resulting in a kerogen fairly poor
195 in H (Type II).

196 In these organogenic layers, the data aforementioned, associated to the high values of
197 S indicate that the partial aerobic biodegradation of the organic matter in its movement along
198 the oxic water column is associated to the selective biodegradation of elements more
199 metabolizable (H) by sulphate reducing bacteria. At specific levels it was identified euxinic
200 environment in peaks of R_{ins} reduction (increase in the carbonate level) not common among
201 the bituminous shales, along with high values of S (Fig. 6). With the prerequisite of sulphate
202 availability dissolved in the water the metaboloc activity of the bacteria promotes the sulphate
203 reduction using the organic matter and generating bicarbonate and sulphidric acid, whose
204 presence characterize euxinic environments. This process of sulphate reduction raises the pH
205 and causes the precipitation of calcium carbonate. Data from biomarkers and isotopes confirm
206 the bacterial influence in the organic matter (Rodrigues et al., 2010a; Rodrigues et al., 2010b;
207 Alferes et al., 2011).

208 In relation to the points characterized as type III (IH<100) and transitional II-III, these
209 occur in lower values of TOC (<2%) (Fig. 6), in fine siliciclastic facies (OR2 and OR4). In
210 these non-organogenic strata the low IH indicates mixture of different sources (both marine
211 and continental). The confirmation of this trend occurs by the predominance of allochthonous
212 palynomorphs (Araujo, 2001) and the correspondence to chemostratigraphic units which
213 indicate marine and terrestrial organic matter according to the biomarkers data (Rodrigues et
214 al., 2010a; Rodrigues et al., 2010b; Alferes et al., 2011). These data reinforce the increase of
215 the marine organic matter in relation to the continental one near to the SIM of the Irati A
216 Sequence.

The organic matter data aforementioned are in accordance to the level of oxygenation of the substrate, since the preservation occurs under low levels. The U values of gamma spectrometry identify less oxygenation beyond the levels enriched in organic matter. With the disponibility of the ion U^{6+} soluble in seawater, in low oxygen levels occurs the reduction to U^{4+} and precipitation as insolubles composts of uranium dioxide (UO_2) or hidroxides ($U(OH)_4$) (Lünning and Kolonic, 2003; Bonotto and Silveira, 2006). Levels of autigenic U occur distributed along the studied section (Fig. 7). Besides the peaks of increase in the U curve, the levels are easily seen in the Th/U curves where values near or below 2 indicate anoxia, and in the curve of occurrence of authigenic U, which stresses the negative covariation with the Th (in the form of monazite: resistate detritic mineral that depends of allochthonous influx) (Fig. 7) (Andersson and Worden, 2004).

The S data supply not only information relative to the organic matter, but also salinity. In the carbonatic strata occur very high S values (3.5) associated to low levels of TOC and R_{ins} (Fig. 6). In these cases the increase of S is in accordance with the presence of sulphate (gypsite and anhydrite), typical of evaporitic IR domains. Therefore the S, which has large disponibility as sulphate dissolved in the sea water, becomes enriched in the inner part of the ramp, as herein described associated to the IR and in thin and isolated levels of the MR. The occurrence of salt layers with texture similar to anhydrite nodules, related to carbonatic strata confirm the aforementioned data. These strata are probably correlated to carbonate associated also to marine organic matter and hypersalinity conditions, according to isotope and biomarkers data (Rodrigues et al., 2010a; Rodrigues et al., 2010b Alferes et al., 2011;). The Th decrease, when associated to IR layers confirm the evaporitic conditions, demonstrating the low humidity according to the reduction of continental detritic minerals input.

In relation to the salinity, besides the high levels herein registered, there are regular salinity conditions in other intervals of the studied section. The values of U and S indicate

marine environment due to their infinite availability in the sea water. Th/U ratio with values between 2 and 7 define oxic marine environment. Variation between this range predominate in the succession (Fig. 7). Accordingly, S values even beyond the organogenic layers even euxinic, present positive oscillations along the section (Fig. 7). The comparison of these inferences to biomarkers data in other strata indicate normal salinity for other intervals of the section (Rodrigues et al., 2010a; Rodrigues et al., 2010b Alferes et al., 2011).

4.3 Stratigraphic scheme of depositional sequences

Three depositional sequences have been identified in the studied interval, as previously mentioned by Araújo (2001) and Hachiro (1996). However, a more detailed register of the system tracts is herein proposed.

The Irati Sequence begins with a transgressive system tract (TST) characterized by the succession of the facies OR3 and OR2 (Fig. 4), with reduction of the granulometry representing a relative rise in the sea-level (Figs. 6). The subtle inflexion of the TOC curve towards higher values is in accordance to this trend, since it indicates either a reduction in the oxygenation close to the substrate or the reduction in the siliciclastic input (Fig. 6). Above lies the highstand system tract (HST), demonstrated by the increase o granulometry in the facies OR4, which corresponds to a progradational trend (Fig. 6). The reduction in the accommodation space associated to the smooth morphology of the ramp facilitated the subaerial exposition even during the short relative sea level variations in this sector of the ramp. In some cases it may cause pedogenetic processes.

On the top of this first sequence, where occurs the IR, it is recorded the forced regression system tract (FRST), where the sea-level lowering allowed the development of biotic activity typical of this type of ramp (Fig. 6). These microbialites are well indicated by the low R_{ins}, demonstrating a high level of carbonates and low TOC, and suitable oxygenation

(Fig. 6). The abrupt contact of this level with the subjacent facies characterizes an erosive surface of marine regression (Fig. 6). The subsequent formation of breccias in these deposits demonstrates a relative sea level fall which caused subaerial exposition characterizing, therefore, a sequence boundary (SB) (Fig. 6). Such boundary is identified in all the cores described in the area. The occurrence, yet isolated, of bioturbation characteristic of excavation in consolidated substrate reinforces this interpretation (Buatois et al., 2002).

The Irati B Sequence begins with a lowstand system tract (LST) characterized by the occurrence of MR (Fig. 4E), where repetitive pulses of shallowing limited by adjacent flooding surface are interpreted as lowstand parasequences (Fig. 6). This pattern is in accordance to the R_{ins} curve which oscillates significantly, and some short and isolated peaks of TOC increase (Fig. 6). At the top of this level the presence of sedimentary evidences of oscillatory fluxes and reworking currents, as well as the increase in siliciclastics, indicates the relative sea-level rise, which occur at the end of the lowstand system tract (Fig. 6). The overlain transgressive system tract culminates in the facies OR1 of black shales (Fig. 4A), which has a flooding surface easily identified in the gamma-ray logs and in the geochemical indicators, especially the TOC between 16% and 20% (Fig. 6). This relative sea-level indicates the restablishment of the OR (Fig. 6). The subsequent highstand system tract is characterized by the reduction of the organic matter content in the shales, easily seen in the TOC in the facies OR2 (Fig. 6). As observed in the previous sequence the contact with the IR is abrupt and is again established an erosive surface of marine regression (Fig. 6). The forced regression system tract is again characterized by the occurrence of bioherms. Subsequently, the IR suffered subaerial exposition resulting in breccias, characterizing a sequence boundary. It is important to mention that in the Irati B Sequence this system tract is not so expressive, being thinner and absent in some cores.

290 The Irati C Sequence has in the base a lowstand system tract again characterized by
291 the the occureence of the MR, howevere thinner than in the previous sequence. Its carbonatic
292 sedimentation is evidenced in the R_{ins} curve (Fig. 6). The adjacent increase in the sea-level
293 rise characterize the transgressive system tract. This tract is demonstrated by the succession of
294 of OR4 and OR1 facies (Fig. 4D and 4A), with the predominance of the outer ramp, besides
295 the trend in the increase in the TOC values (Fig. 6). The reduction in the organic matter
296 amount in the subsequent unit (Serra Alta Formation), characterize the highstand system tract
297 (Fig. 5).

298 It is important to mention that previous studies have already made allusion to the
299 climatic control of facies succession in the Irati Formation, ascribing a 4^{th} order hierarchy to
300 the ciclicity herein observed in the three depositional sequences (Araújo, 2010; Hachiro,
301 1996).

302 4.4. High frequency cycles

303 Cyclic transgressive-regresive (TR) internal oscillations have been observed along the
304 three depositional sequences (Fig. 7) according to Embry and Johannessen (1992). The
305 cyclicity was observed in the total curve of the gamma-ray log, whose radioactivity
306 corresponds to the sum of K, Th and U (Glover, 2012). In the case of siliciclastic rocks of fine
307 granulometry that represents the highest part of the succession herein described, the variations
308 of K are associated to the clay content of the rocks, indicating variations of higher and lower
309 energy in the environment due to oscillations in the water depth (Milani, 1997; Kearey et al.,
310 2009). Therefore, it is assumed that the increase in the K values correspond to a transgressive
311 trend. The detritic Th as mineral monazite resistates (Andersson and Worden, 2004; Kearey et
312 al., 2009), presents negative covariation with K, indicating higher energy intervals and
313 increased sedimentary input. On the other hand, in the cases of positive covariation with K,
314 the Th is associated to clay minerals by adsorption (Syed, 1999). In the case of U, due to its

behavior in relation to other radioactive elements, presenting negative covariation, it was considered as authigenic, corresponding to low oxygenation contexts (as previously detailed).

The total amount of gamma ray is defined predominantly by the K, due to its relative abundance in relation to the other radioactive elements measured. However, it is influenced by the U and the Th, as negative covariations, reducing the effect of K in the total curve, as well as positive covariation, which reinforce the effect of K in the curve.

Positive covariation was observed in eight out of nine analyzed cycles in the succession, influenced predominantly by the K variation. Due to the affinity of U with low oxygenation levels, such covariation suggests oxygen depletion in transgressive trends. The increase peak of K with a negative covariation of U in the cycle eight demonstrates a rise in the relative sea-level, without the establishment of low oxygenation context.

Besides the transgressive peaks indicated by K, there are also significant increase in the U values that do not coincide with the K peaks and, therefore, are not conspicuous in the total curve. These levels, which indicate low oxygenation, are not related with the sea level rise. It was not observed covariation between U with TOC to justify the oxygen depletion by events of productivity increase. Therefore, the peaks of authigenic U indicate climatic changes which influenced the hydric imbalance and the circulation patterns.

As mentioned in the previous sub-item, the climate is the major controller of sequences deposition in the 4^{th} order scale (Araújo, 2010; Hachiro, 1996). Besides, it is possible to infer that the cycles of highest frequency herein described, which are also mostly influenced by the climate, are of 5^{th} order hierarchy.

5. Final considerations

The depositional system of the Irati Formation was interpreted as a carbonatic ramp with occurrence of three domains: inner, middle and outer. The succession of the domains is

controlled by the relative sea level changes which correspond to three depositional sequences with TST, HSST, FRST and LSST. The depositional dynamics is complex and variable, since it is influenced not only by oceanic incursions, but also the climate and, consequently the continental input.

Along the section there are variations in the salinity and oxigenation of the substrate. In general terms, there are predominance of normal salinity in the fine siliciclastic facies of the OR both in the organogenic and in non-organogenic ones. In the carbonatic facies of the IR and MR there are evidences of high salinity. There is also variations in nthe composition of the organic matter. In the carbonatic facies of the IR and the MR the organic matter is of marine origin. In the fine siliciclastic non-organogenic facies there are predominance of marine organic matter, however with continental input. In the organogenic the precursors of the organic matter are of bacterian origin.

This indicates different conditions of hydric imbalance. In two moments of the sucession there was the establishment of an evaporitic environment in a semi-arid climate, with low humidity and poor continental contribution. Such conditions are demonstrated in the IR domain, associated to the RFST, where took place a microbial carbonatic sedimentation under oxic conditions, in an environment of high salinity and subaerial exposition.

In the external ramp domain, associated to the TST developed na anoxic and euxinic environment with normal salinity. This paleoenvironmental context resulted from the relative sea level rise and increase in the productivity during phases of higher humidity.

It can be inferred that the factors that control the sedimentation are linked to phenomena of 4^{th} order. Near to the maximum flooding surface, the reduction of the continental input demonstrates the predominance of allochthonous control (oceanic influx) in the sea level oscillation in this sector of the basin in relation to the autochthonous control

(continental contribution). The 4th order ciclicity defines the relative sea level by eustatic variations and climatic influence, as seen in oscillations between periods of low and high humidity.

In relation to the high frequency cycles were observed T-R cycles identified in the oscillations of the K amount, highlited by the U increase. This indicates lower oxygenation near to the substrate according to the transgressive trend. Besides the T-R cycles, the peaks of U increase indicate phases of lower oxygenation beyond the transgressive trend. In these cases of high frequence oscillation, the absence of relation with the TOC demonstrates the climate a the controlling factor of the cycles.

In the case of oxygen depletion as evidenced by the absence o U in the transgressive peak the climate influences the deposition by the interference in the hydric imbalance between the continental input and the marine influx. This promotes the increase or reduction of the bottom circulation due to the intermittent presence of thermohaline circulation, which reduces the oxygen supply toward the susbstrate. In the case of T-R cycles evidenced by K the climatic influence can even cause the increase in the thickness of the water column during the transgressions.

It was noticed cyclicity in the deposition of the Irati Formation through the depositional sequences and T-R cycles of highest frequency. Its hierarchy corresponds to cycles of 4th and 5th order, respectively.

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Figure captions

Fig. 1. Simplified geologic map of the Paraná Basin showing the structural outline of the basement, units and distribution of the supersequences described in the text (adapted from Milani, 1997).

Fig. 2. Stratigraphic diagram of the Gondwana I Supersequence, showing the 2nd order stratigraphic surfaces (adapted from Milani, 1997). In evidence is the level of Irati Formation. *CB – Corumbataí Formation, whose age is equivalent to the Serra Alta and Teresina formations.

Fig. 3. Geographic localization of the study area with the lithoestratigraphic unit and the cores descripted.

Fig. 4. Photos of the facies described and commented along the text. A) Facies OR1 of black shales. B) Facies OR 2 of deep gray argillaceous-siltstone with fissility and incipient lamination. C) Facies OR3 of light gray siltstone. D) Heterolithic Facies OR4. E) The OR level is in dashed, overlain by the MR in continuous line. F) Fossiliferous level with fish sacales of the OR2 facies. G) Fossiliferous level with *Mesosuarus* remains of the OR4 facies. H) Tepee of MR shallowing. I) Salt layer similar to anhydrite nodules in the MR.

Fig. 5. Left: Correlation graphics of the oxygen index (IO) and hydrogen index (IH) of two probing and their resulting kerogen compositional fields. Right: Correlation graphics o total

organic carbon (TOC) and IH, a standard line demonstrating the asymptotic behaviour in the cluster.

Fig. 6. Lithologic and stratigraphic profile of two described cores showing the depositional sequences, their system tracts, the domains of the carbonatic ramo and the curves resulting from the geochemical analyses of the total organic carbon (TOC), sulphur (S), hydrogen index (HI) and insoluble residue (R_{ins}).

Fig. 7. Stratigraphic and lithologic profiles of the core described with gamma spectrometry, showing the total amount curve of gamma ray and the differentiable channels (U, Th, K), and derivative ratios. In the gamma ray curve (total) the cycles T-R are highlighted.

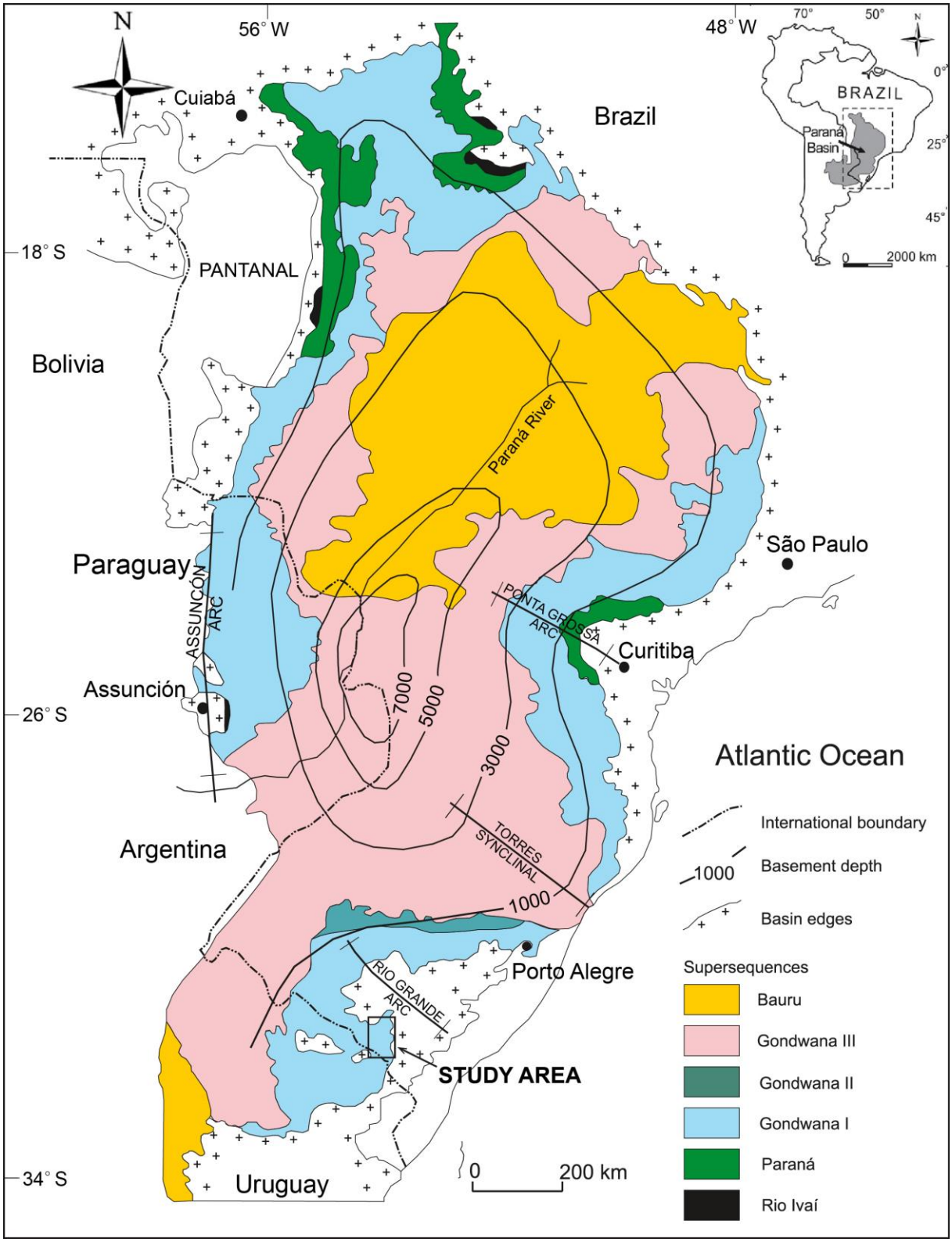
N
56° W
48° W
Cuiabá
Brazil
BRAZIL
Paraná Basin
70°
50°
0°
25°
45°
0
2000 km
18° S
PANTANAL
Bolivia
Paraná River
São Paulo
Paraguay
ASSUNCIÓN ARC
PONTA GROSSA ARC
Curitiba
Assunción
26° S
7000
5000
3000
Atlantic Ocean
TORRES SYNCLINAL
Argentina
1000
International boundary
1000
Basement depth
Basin edges
Supersequences
Bauru
Gondwana III
Gondwana II
Gondwana I
Paraná
Rio Ivaí
Porto Alegre
RIO GRANDE ARC
STUDY AREA
34° S
Uruguay
0
200 km

Fig. 1

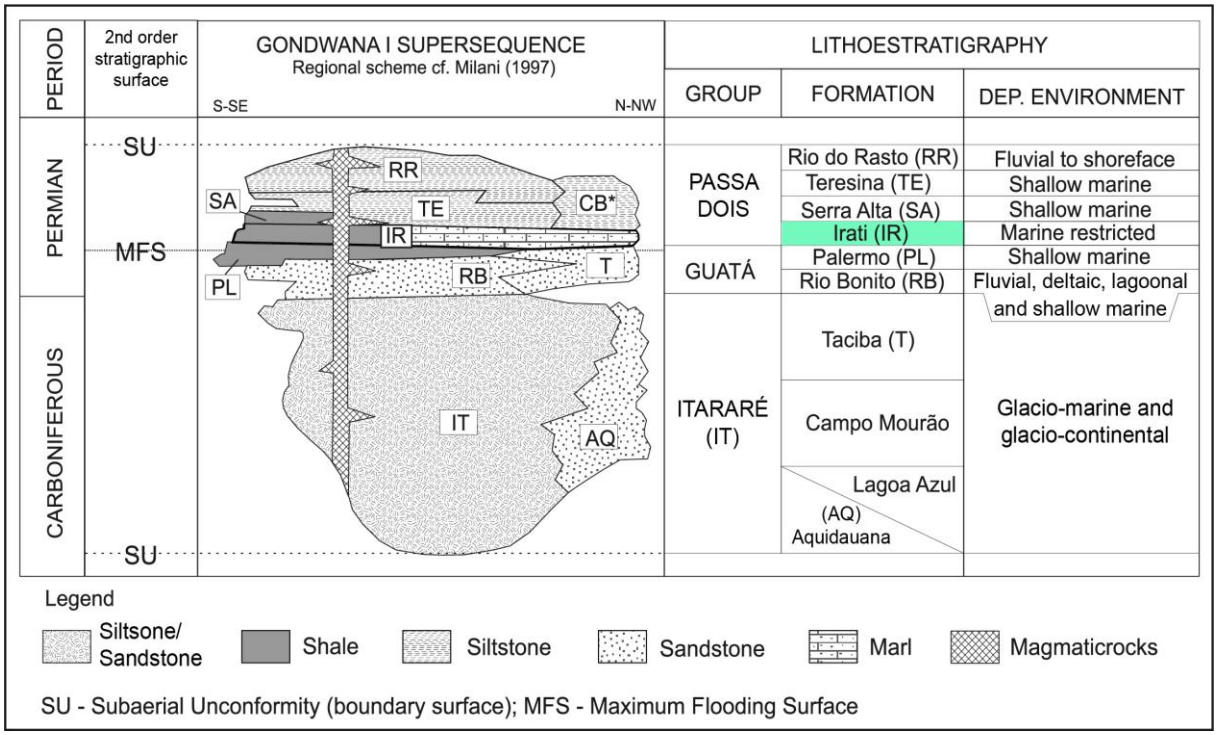
PERIOD
2nd order stratigraphic surface
GONDWANA I SUPERSEQUENCE
Regional scheme cf. Milani (1997)
S-SE
N-NW
LITHOESTRATIGRAPHY
GROUP
FORMATION
DEP. ENVIRONMENT
PERMIAN
CARBONIFEROUS
SU
MFS
SU
SA
PL
RR
TE
IR
RB
CB*
T
IT
AQ
PASSA DOIS
GUATÁ
ITARARÉ (IT)
Rio do Rasto (RR)
Fluvial to shoreface
Teresina (TE)
Shallow marine
Serra Alta (SA)
Shallow marine
Irati (IR)
Marine restricted
Palermo (PL)
Shallow marine
Rio Bonito (RB)
Fluvial, deltaic, lagoonal and shallow marine
Taciba (T)
Campo Mourão
Glacio-marine and glacio-continental
Lagoa Azul
(AQ)
Aquidauana
Legend
Siltsone/ Sandstone
Shale
Siltstone
Sandstone
Marl
Magmaticrocks
SU - Subaerial Unconformity (boundary surface); MFS - Maximum Flooding Surface

Fig. 2.

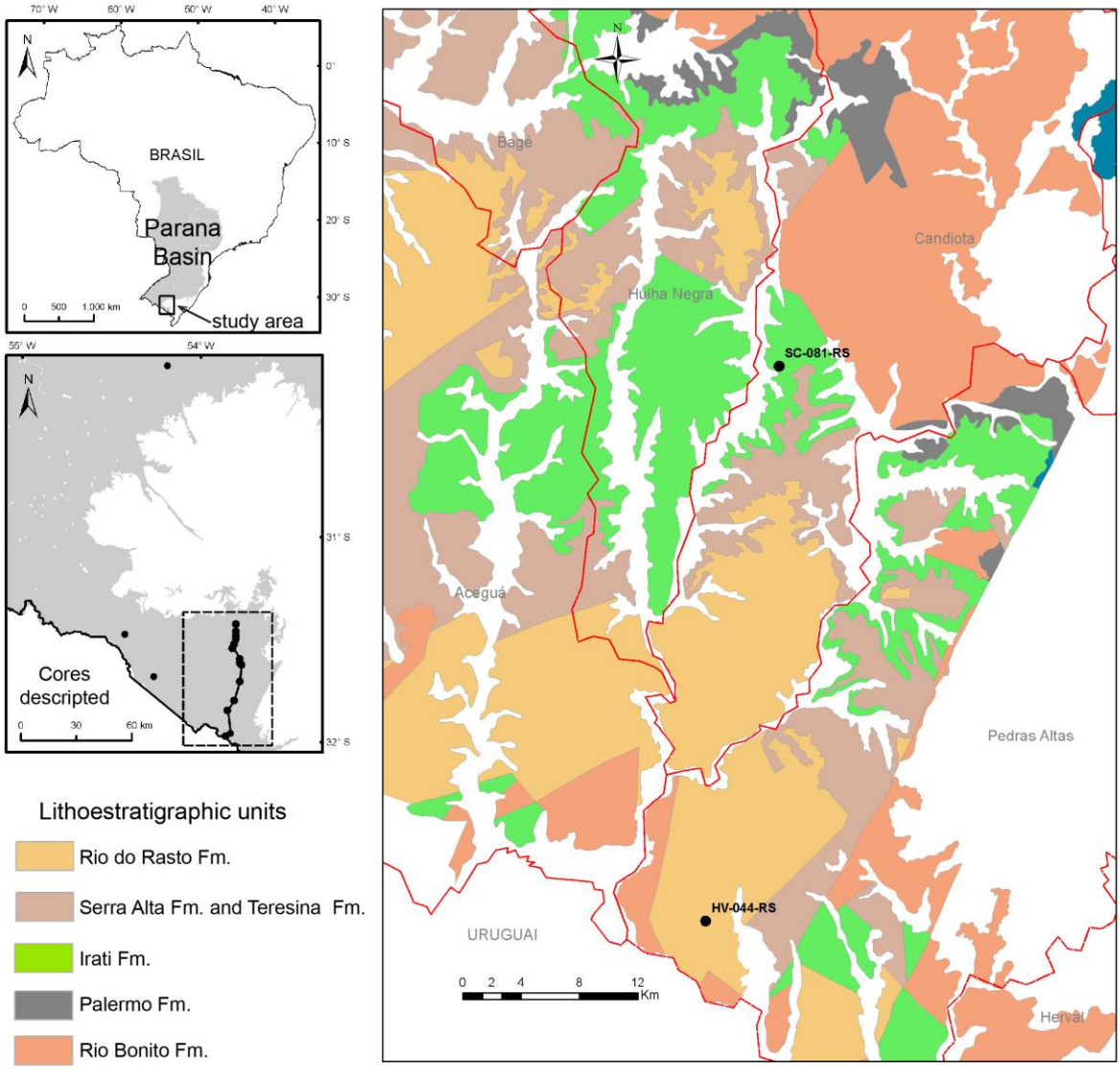
70° W
60° W
50° W
40° W
0°
10° S
20° S
30° S
N
BRASIL
Parana
Basin
study area
0 500 1.000 km
55° W
54° W
31° S
32° S
Cores
descripted
0 30 60 km
Lithoestratigraphic units
Rio do Rasto Fm.
Serra Alta Fm. and Teresina Fm.
Irati Fm.
Palermo Fm.
Rio Bonito Fm.
Bagé
Candiota
Hulha Negra
SC-081-RS
Aceguá
Pedras Altas
HV-044-RS
URUGUAI
0 2 4 8 12
Km
Herval

Fig. 3

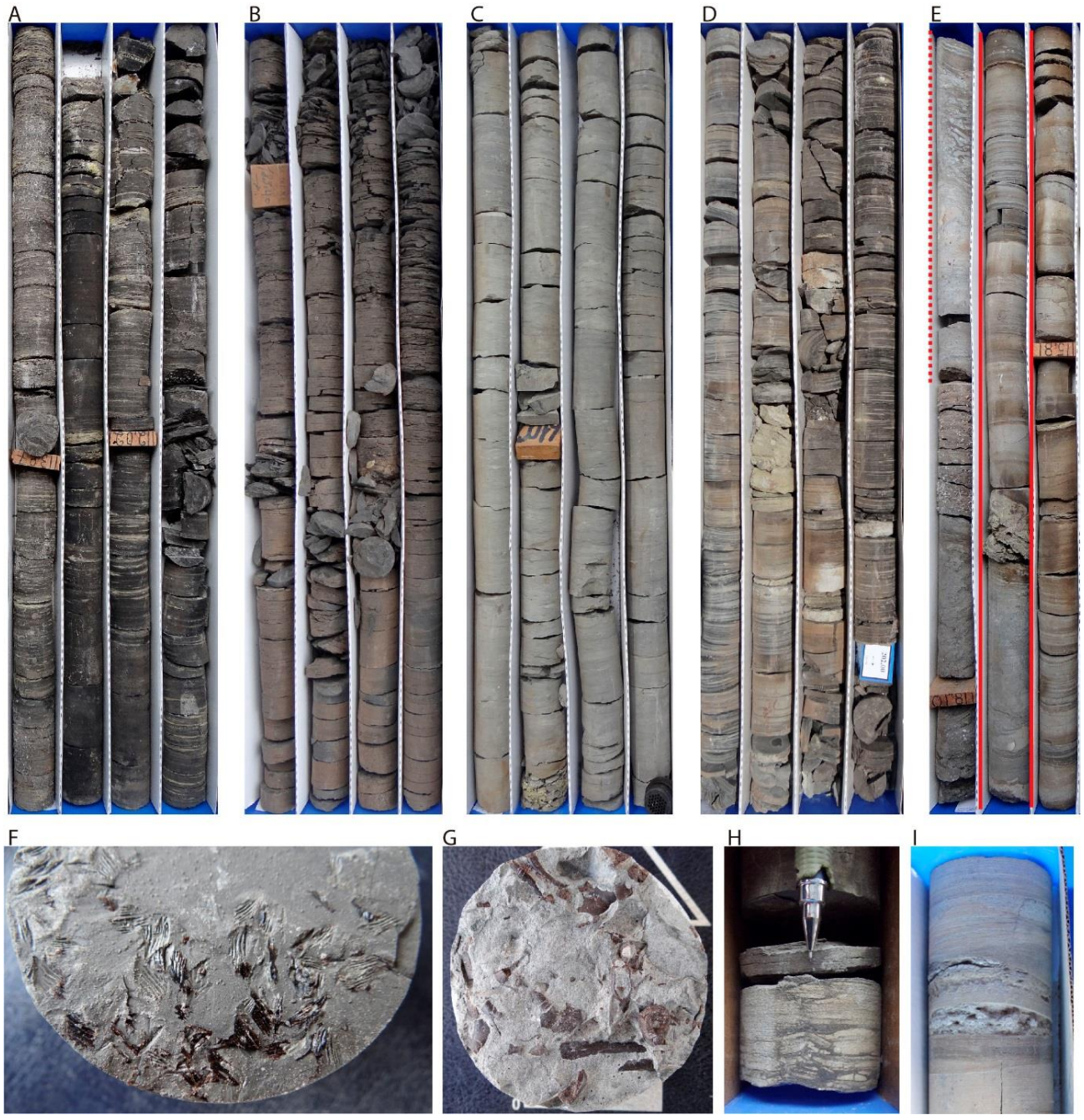

Fig. 4.

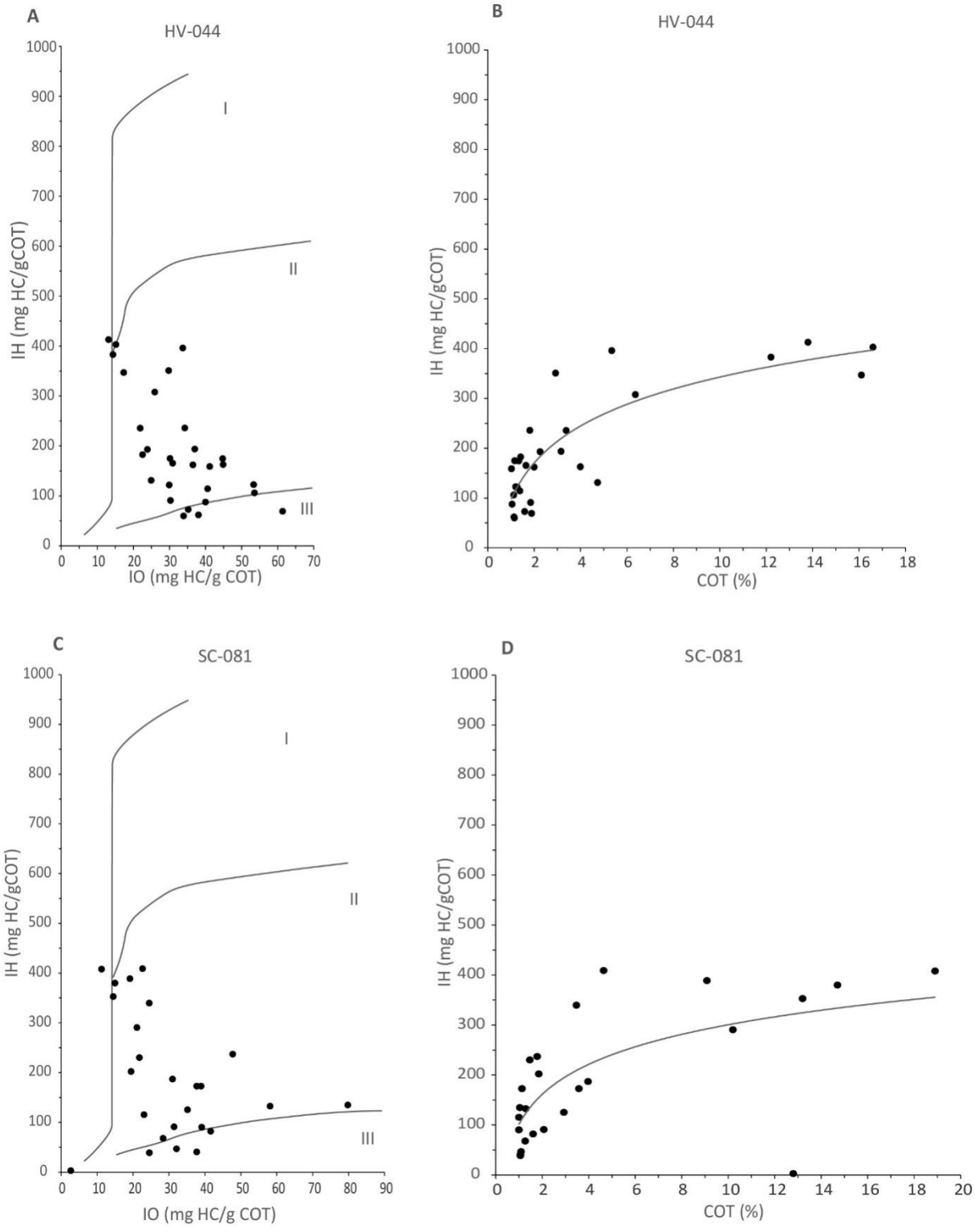
A
HV-044
IH (mg HC/gCOT)
IO (mg HC/g COT)
I
II
III
B
HV-044
IH (mg HC/gCOT)
COT (%)
C
SC-081
IH (mg HC/gCOT)
IO (mg HC/g COT)
I
II
III
D
SC-081
IH (mg HC/gCOT)
COT (%)

Fig. 5.

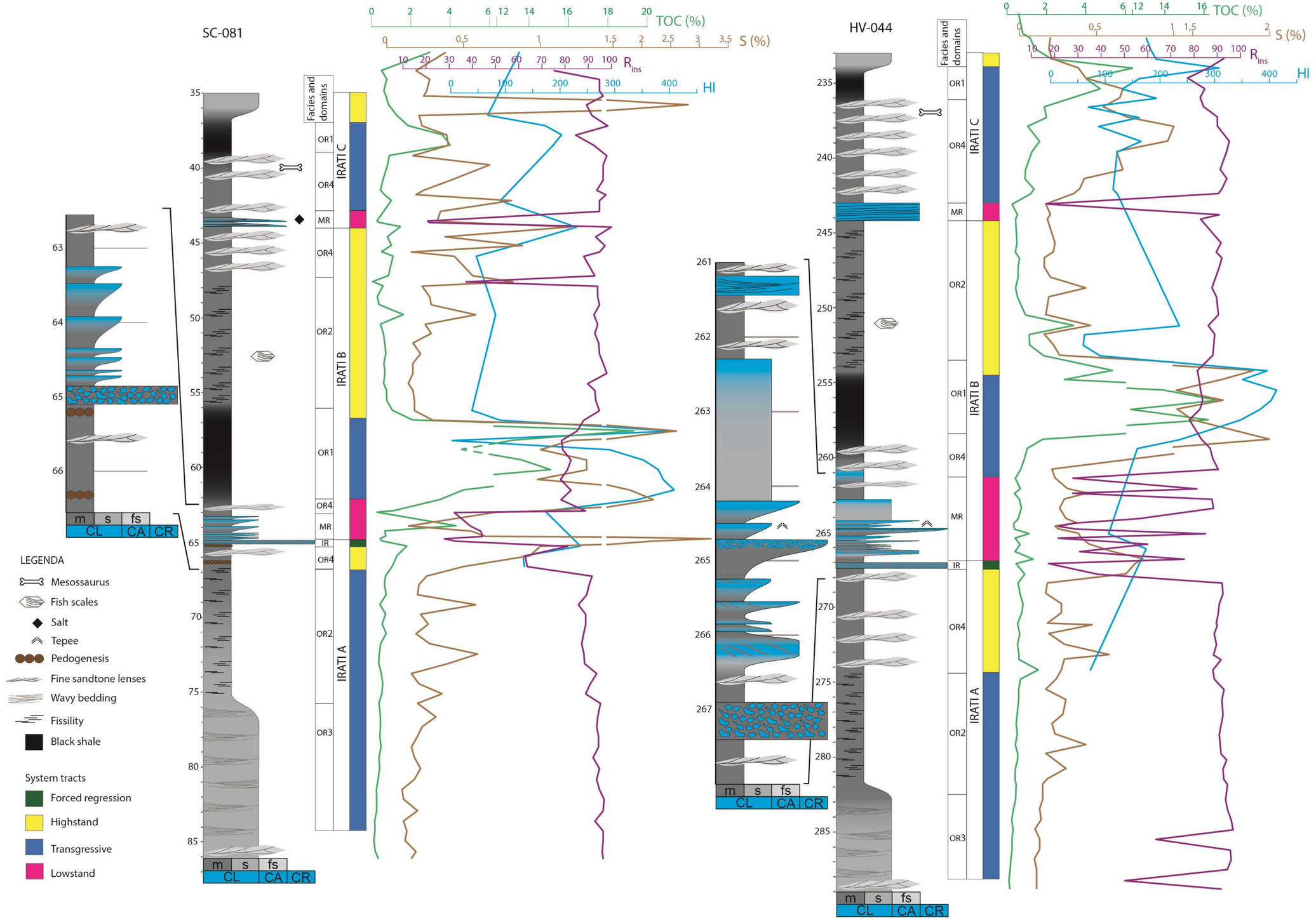
SC-081
HV-044
TOC (%)
S (%)
R_ins
HI
Facies and domains
OR1
OR4
MR
IR
OR2
OR3
IRATI C
IRATI B
IRATI A
m
s
fs
CL
CA
CR
LEGENDA
Mesossaurus
Fish scales
Salt
Tepee
Pedogenesis
Fine sandtone lenses
Wavy bedding
Fissility
Black shale
System tracts
Forced regression
Highstand
Transgressive
Lowstand

Fig. 6.

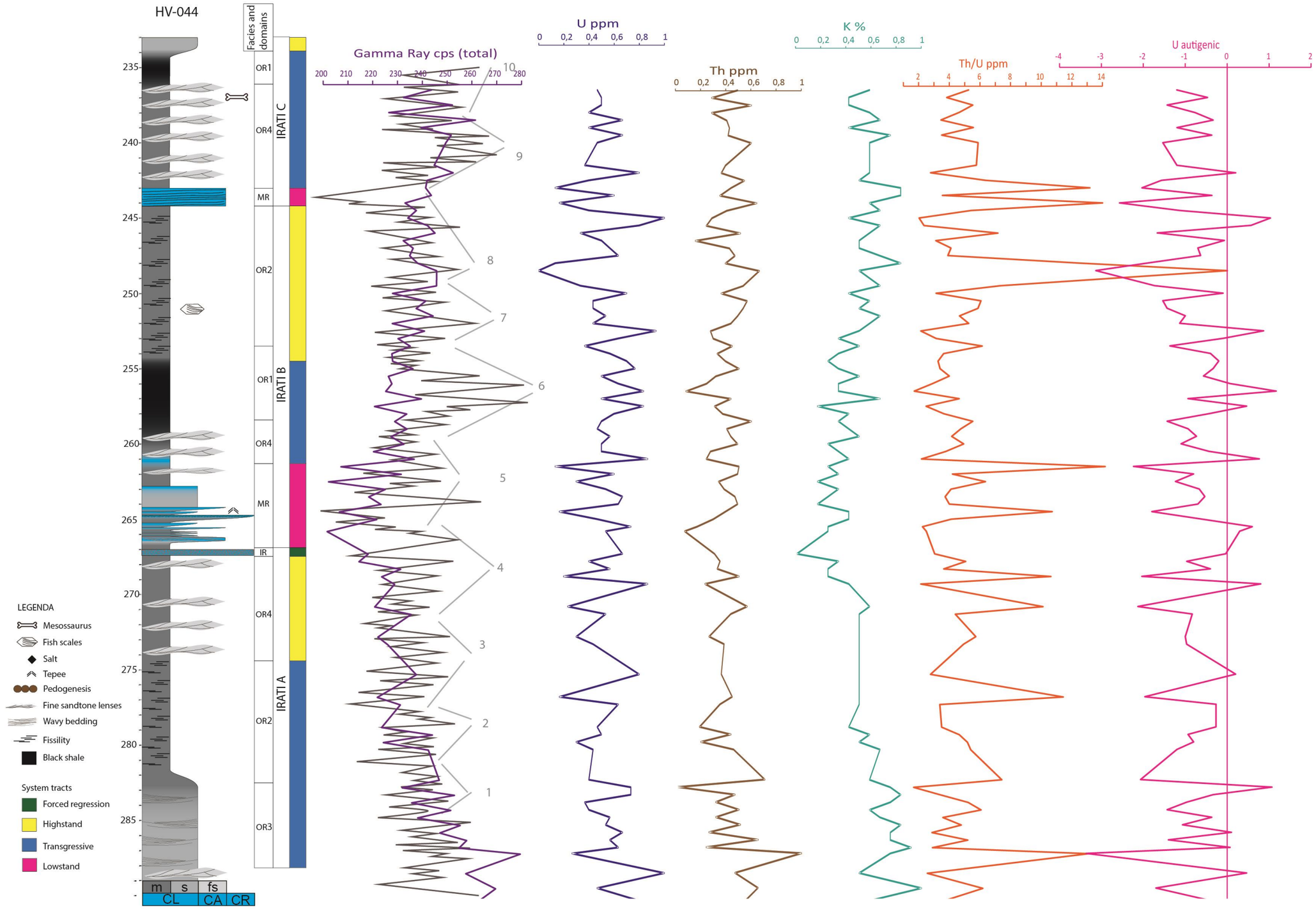
HV-044
Facies and domains
OR1
OR4
IRATI C
MR
OR2
OR1
IRATI B
OR4
MR
IR
OR4
IRATI A
OR2
OR3
235
240
245
250
255
260
265
270
275
280
285
Gamma Ray cps (total)
200 210 220 230 240 250 260 270 280
U ppm
0 0,2 0,4 0,6 0,8 1
Th ppm
0 0,2 0,4 0,6 0,8 1
K %
0 0,2 0,4 0,6 0,8 1
Th/U ppm
2 4 6 8 10 12 14
U autigenic
-4 -3 -2 -1 0 1 2
10
9
8
7
6
5
4
3
2
1
LEGENDA
Mesossaurus
Fish scales
Salt
Tepee
Pedogenesis
Fine sandtone lenses
Wavy bedding
Fissility
Black shale
System tracts
Forced regression
Highstand
Transgressive
Lowstand
m
s
fs
CL
CA
CR

Fig. 7.

SÍNTESE INTEGRADORA

As inferências paleoambientais discorreram acerca da alternância de períodos úmidos e semi-áridos, associados a oscilações entre concentração e diluição salina, e oxigenação do substrato, que ocorreram ao longo do empilhamento das fácies de folhelhos, folhelhos betuminosos e carbonatos. Este resultado constituiu o contexto final do trabalho. Neste contexto também foi feita a caracterização da matéria orgânica presente na Formação Irati, a qual se configura de forma heterogênea como marinha, bacteriana e até com níveis continentais. Para chegar a tais conclusões foi feito uso de ferramentas de geoquímica orgânica. Por outro lado, também no contexto mais adiante do presente trabalho, foi possível problematizar as hierarquias da ciclicidade presente nas sequencias deposicionais e nos ciclos T-R, correspondendo a quarta e quinta ordem, respectivamente. Esta etapa contou com os dados detalhados de gamaespectrometria.

Para acessar esta instância de resultados, antes do ferramental analítico de laboratórios, foi necessário o domínio do arcabouço faciológico e estratigráfico da Formação Irati no setor sul da Bacia do Paraná. Tal etapa só foi possível por meio da descrição de um considerável numero de testemunhos de sondagem, bem como correlação com perfis geofísicos de outras sondagens para obter o controle lateral das fácies. Fruto desta etapa foi a caracterização do sistema deposicional como rampa carbonática, composta pelos domínios externo, médio e interno. A análise do empilhamento das fácies permitiu delimitar três sequencias deposicionais, que compuseram a posterior problematização das hierarquias.

Desta forma, por meio desta síntese integradora nota-se que os resultados alcançados foram capazes de contemplar o que inicialmente foi proposto nos objetivos. O cenário paleoambiental da Formação Irati no sul da Bacia do Paraná, apresentado neste estudo, pode alimentar as discussões acerca da dinâmica ambiental do Permiano.