Coal petrology analysis and implications in depositional environments from Upper Cretaceous to Miocene: a study case in the Eastern Cordillera of Colombia

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Abstract

Coal petrological characteristics along the Piedemonte Llanero and the reconstruction of the deposit environment were obtained from macerals and micro-lithotypes analysis since these data provide information about the processes and prevalent conditions during the peat formation. We analyzed seams from Cenomanian to Miocene geological units (Chipaque Formation, Palmichal Group, Arcillas del Limbo Formation, and San Fernando Formation). Coal range decreases gradually from high-volatile C bituminous (HVCB) in the Chipaque Formation to sub-bituminous C in the San Fernando Formation. The coals are enriched in macerals of vitrinite, whereas the liptinite and inertinite concentrations vary according to the stratigraphic position. The micro-lithotypes are bi-maceral and tri-macerals, being the highest concentrations of clarite and vitrinertoliptite. The results of the facies analysis show that the peat in which the coals developed is mainly of arboreal and herbaceous affinity (rich in lignin and cellulose). Peats are ombrotrophic (rainfed) to mesotrophic (transitional or mixed mires) with variations in the flooding surface and influxes of brackish water. Good tissue preservation is inferred from the wet conditions in forest swamps with few humification and gelation. According to the micro-lithotypes composition, the peat environment was deduced as estuarine system, evolving to lacustrine environment of the deltaic system, both restricted by changes in sea level, which are evidenced by oxic and anoxic periods in the analyzed sequence.

Keywords: Eastern Cordillera basin, Northern Andes, Coal, Organic Petrology

Introduction

Coal is one of the main commodities of the Colombian economy, 4,881 million tons were reported as reserves of bituminous and anthracitic coal during 2019. Colombia is the major producer of South and Central America, and is the sixth coal exporter, especially to Europe (BP 2019). Twelve coal zones are recognized along with the country, and the Piedemonte Llanero is one of the least known from the coal resources point of view (Ingeominas 2004). Preliminary studies of the Servicio

Geológico Colombiano have established different stratigraphic levels for coal, from the late Cenomanian to Miocene, as the Chipaque Formation, Palmichal Group, and Arcillas del Limbo and San Fernando formations (Ingeominas 2004; Monroy and Sandoval 2017; Monroy and Patiño 2018). A transitional deposit environment of these geological units was inferred in previous stratigraphy and palynology works (van der Hammen 1958; Ulloa and Rodríguez 1976b; Sarmiento 1992; Guerrero and Sarmiento 1996; Pardo 2004; Dueñas and van der Hammen 2007; Bayona et al. 2007; Bayona et al. 2008; Parra et al. 2008; Jaramillo et al. 2011; Pardo and Jaramillo 2014; Caballero et al. 2020). The Paleocene vegetation was determined as angiosperms, that later, during middle Eocene – Oligocene was predominately pteridophytes – bryofites, under at least five marine incursions determined for the central part of the Eastern Cordillera (Pardo and Jaramillo 2014) to the north of the present study area.

Stratigraphic, sedimentological and palynological studies have been carried out along the Piedemonte Llanero, mainly focused on oil and gas resources, although coal petrology is not considered yet. From a systematic sampling, this technique has been applied in the Cundinamarca Zone, also located in the Eastern Cordillera, where the results showed the paralic transgressive to limno-telmatic environments for the bituminous coals of the Guaduas Formation during the Maastrichtian-Paleocene (Guatame and Sarmiento 2004; Mejía et al. 2006; Gómez and López 2017). Whereas, along the western flank of the Central Cordillera, Castaño and Gómez (2001) carried out a coal petrology analysis for the Antioquia-Antiguo Caldas Zone getting a fluvial environment with possible meandric currents for the low-rank coals of the Miocene-Pliocene Aranzazu volcano-clastic sequence, classifying them as lignite to C sub-bituminous. For the same zone, but in the

Amagá-Nechí sector, Blandón (2007) determined estuarine and lower delta depositional conditions for the sub-bituminous coals of the Amagá Formation, whose age is uncertain, although a Late Oligocene – Early Miocene has been dated from palynology. Whereas, in the Los Cuervos Formation from the Calenturitas and La Jagua coal mines in the Cesar zone, Guo et al. (2018) determined ombrogenic conditions without any major marine influence during most of the time of peat deposition are also deduced of sub-bituminous coals.

We want to demonstrate the importance of integrated data from coal petrology, palynology, and sedimentology of associated sedimentary rocks. These disciplines are essential to support and restore the deposit environment and infer the origin of vegetal matter, facies, processes, and conditions of the coal formation. Petrology provides indices and ternary diagrams as elements for this sort of interpretations (Teichmüller and Teichmüller 1982; Diessel 1986, 1992; Mukhopadhyay 1986; Calder et al. 1991; Singh and Singh 1996; Singh and Singh 2000; Rimmer et al. 2000; Staub 2002; Singh et al. 2013; Bechtel et al. 2014; Sen et al. 2016; Singh 2016). Some authors argue the disagreement on the use of diagrams and indexes (Dai et al. 2020). However, we consider that the models apply the fundamentals related to the active processes at the time of peat formation, the appearance and nature of the maceral groups, and thus we include them in our analysis.

The merge of the coal petrology results with other already named disciplines offers more convincing and supported interpretations in different scopes to improve the geological knowledge, optimize the use of the resources, and the exploration of potential gas zones for domestic consumptions, considering the shortage prognostic in Colombia. This is related to the coal quality, in which petrology helps to define the type of coal (nature of the organic components), grade (extension of the mineral matter), and rank. Basin analysis supported by coal petrology provides elements to infer the deposit environment of the organic material, the thermal history, the type, and the potential of the hydrocarbons source rock. Additionally, coal petrology is a fundamental tool in the survey of new alternatives for energy generation as coal-bed methane (CBM), by helping in the discernment of macerals that contribute to gas adsorption and desorption capacity, as well as in the necessity of raising the knowledge and rare earth elements concentration in coal seams.

Geological frame

The Piedemonte Llanero is located in the eastern foothills of the Eastern Cordillera Basin, which was configured in an asymmetric graben under marine conditions near the coast with an alternation to continental deposits; and besides affected by different events that caused tectonic inversion, especially from the early Eocene to Oligocene (Barrero et al. 2007). The main regional structures (SW- NE) in the area are the Boquerón, Recetor, Porvenir, Nazareth, and Río Amarillo synclines, located between the thrust fault system of Tesalia to the west and Guaicáramo to the east (Fig. 1).

The coal formations correspond in age from the Upper Cretaceous to the Neogene. The sedimentary sequence began with the Chipaque Formation (Upper Cenomanian -Santonian in Terraza et al. 2013) and consists of shale and claystone with thin sandstones interbedded, which, towards the top, become more frequent and are observed in more than 2 m thick, with sets up to 25 m. Seven coal seams are identified in the lower part of the formation, numbered M101 at the base and M107 at the top. The seams thicknesses are 0.4 to 1.80 m (Monroy and Patiño 2018). For the Maastrichtian-Lower Palaeocene interval, the nomenclature of the Palmichal Group (Ulloa 1976b) was adopted in this paper, although different authors have found controversial given its equivalences with other formal units as Guadalupe Group and Guaduas Formation (Guerrero and Sarmiento 1996; Terraza et al. 2013). The base of the Palmichal Formation consists of three thick levels of white, medium-grain, and good selection sandstones in medium to very thick beds, with intercalations of grey to dark-grey claystone and siltstone levels, and thinner levels of medium-grained sandstone in thin to medium beds. Towards the top, this formation comprises banks of yellow, medium to coarse-grained sandstones with poor selection and quartz crystals reaching 5 mm in diameter (Monroy and Sandoval 2017). The thicknesses of the coal seam vary between 0.40 and 0.92 m in the sector of Jagua-Humea (Monroy and Patiño 2018). The age of the formation is Early Campanian to Paleocene (Terraza et al. 2013). The Paleocene comprises the Arcillas del Limbo Formation (Van der Hammen 1958, 1960; Monroy and Sandoval 2017), a sequence characterized by grey, reddish, and green claystone, and a thick succession of white and yellow sandstones bands. Additionally, there are coal-ribbons and seams that are stratigraphically located in the lower and middle sections of the sequence. The M201, M202 seams vary in thickness between 0.4 to 0.8 m and 0.9 to 1.6 m, respectively; and for the coal-ribbons vary between 0.15 to 0.35 m (Monroy and Sandoval, 2016). The San Fernando Formation was dated from the late Eocene-Oligocene (van der Hammen, 1958), or Middle Miocene for the upper segment according to Dueñas and van der Hammen (2007), who correlate it with the León Formation of the Catatumbo Basin. Furthermore, the unit is coeval with the Carbonera Formation, a name given by the oil companies and divided among eight members, from C1 to C8 (Bayona et al. (2007, 2008); Parra et al. (2008); Pardo and Jaramillo (2014); Caballero et al. (2020). The formation consists of grey claystone with sandstones of medium to coarse grains interbedded in banks up to 3 m thick. Five coal seams (M301 to M305) were defined from 0.4 to 1.1 m of thicknesses (Monroy and Sandoval 2017) Fig. 2.

Methodology

A total of fifteen coal seams of the Piedemonte Llanero were analyzed. The proximate analysis (moisture, ash, volatile matter, and fixed carbon), ultimate analysis (C, H, N, S, and O) and measurement of calorific value were performed to determine the rank of coal, according to the international standards (ASTM D388-99 (1999), ISO 11760-2005). From the percentages of C, H, and O elements the atomic radius was calculated, and results were plotted in the Van Krevelen diagram (Tissot and Welte 1978) to identify the type of kerogen.

The microscopic analysis included reflectance measurement (% Ro). The quantitative and qualitative observation of the macerals, under reflected light and fluorescence, were performed according to the classification given in the compilation of ICCP (1963, 1971, 1975, 1998, 2001); Sýkorová et al. (2005) in Suárez and Crelling (2008). The procedure is standardized as indicated in the ISO 7404/3 (2009), ASTM D2799-05a, and D2798-05b norms. The micro-lithotype analysis is carried out on a 20-intersection reticule as indicated in the ISO 7404/4 (1988) norm, and the classification given in ICCP (1963, 1971), in Suárez and Crelling 2008) and Taylor et al. (1998).

From the petrographic results, models, and indices (GI, TPI, GWI, VI) proposed by different authors in recent decades were used to identify the facies and deposit environment of the coals (Smyth 1979; Mukhopadhyay 1986; Calder et al. 1991; Diessel 1992; Singh and Singh 1996; Rimmer et al. 2000; Singh and Singh 2000; Singh et al. 2013). Regarding the current knowledge about the stratigraphy and sedimentology of the Eastern Cordillera, different studies were integrated to analyze our results, we took into consideration mainly the outcomes of Ulloa (1976), Guerrero and Sarmiento (1996), Dueñas and van der Hammen (2007), Bayona et al. (2007, 2008), Parra et al. (2008), Terraza et al. (2013), Pardo and Jaramillo (2014), and Caballero et al. (2020).

Results

Coal rank and type of kerogen

The results of the huminite- vitrinite reflectance and the proximate analysis (moisture, volatile matter, fixed carbon, and ash) on an ash-free dry base were required. It was observed that the rank of coal decreases as the units got younger. Towards the base the coals of the Chipaque formation are classified as BAV (M101-M107), followed by the seam of the Palmichal Group (M151), categorized as BAVC. The seams of the Arcillas del Limbo formation were ranked as subbituminous A (M201- M202), and those of the youngest San Fernando formation as subbituminous A, B, and C- M301- M305 (Table 1).

The diagram of Van Krevelen (1961, 1993) is based on the content of H, C y O, referring to diverse forms of organic matter as a precursor of petroleum (Kerogen). The kerogen is classified by the chemical composition mainly, it is related to the coal macerals as indicators of the original organic matter and the environment depositional, providing elements of the physical and biological processes that occurred during burial (van Krevelen 1961; Cornelius 1978; Robert 1980; Stach et al. 1982).

From H/C, O/C atomic ratio the coal seams analysed (Fig. 3) are Type III, characteristic of humic organic matter, derived from continental plants with identifiable detritus and deposited in proximal environments (Tissot and Welte 1978; Suárez et al. 2012), also associated with a lacustrine or marine environment with a terrestrial influence (Suárez et al. 2012). The atomic ratios allow us to identify the degree of carbonization. This analysis showed that increasing the oxygen concentration decreases the coal rank, while the percentage of hydrogen indicates the type of kerogen (Fig. 3).

Maceral and micro-lithotype analysis

Huminite- Vitrinite is the dominant group of macerals, showing a gradual increase in its percentage from the Palmichal Group to the San Fernando Formation. Macerals of the liptinite group are found in higher percentages in the middle section of the stratigraphic column (Arcillas del Limbo Formations and Palmichal group), and the percentage of macerals of the inertinite group is higher in the coals of the Palmichal group (Fig. 4; Table 2).

Chipaque Formation

The Table 3 and Fig. 5 show the quantitative distribution and photographs of macerals of the Chipaque Formation, in which coals are mainly vitrinitic, with the telovitrinite as the dominant subgroup, whereas the collotelinite is the maceral that predominates in most seams, except in M102, in which gelification becomes more evident with the increase in the percentage of the subgroup of gelovitrinite-corpogelinite (17.76% vol). Liptinite, with average values not lower than 13.68% vol, is represented by sporinite, cutinite, and liptodetrinite (M103). The group of inertinite has a minimum value of 18.05% vol. The inertodetrinite, fusinite, semifusinite, and micrinite are the most abundant macerals. The mineral matter, with a maximum value of 13.20% vol in M104, is composed of clay, carbonate, and pyrite.

Vitrinite group. The telinite maceral occurs associated with corpogelinite, it is filling bodies with rounded and oval shapes. The collotelinite is found as thick bands, while the collodetrinite appears as a groundmass that contains different preserved and crushed macerals. Occasionally, cracks and marks of the oxidized coal are observed.

Liptinite group. The sporinite was observed as a microsporinite mainly aligned with the stratification. A lower proportion of macrosporinites, pollen, and sporangium are also found. Thin walled cutinite (tenuicutinite), serrated edges and thick-walled cutinite (crassicutinite) deformed has been identified. Resinite occurs filling cracks or empty spaces or replacing other macerals, it exhibits round to irregular shapes and a variable size, immersed in collotelinite, it has a darker grey colour. The chlorophyllinite, derived from the chloroplasts with red fluorescence (Pickel et al. 2017), is observed in a low proportion and reveals strongly anaerobic conditions in coals formed from forest peat. Bituminite appears as amorphous or a sub-rounded- elongated groundmass occasionally attached to other macerals, and another secondary maceral is exudatinite, occurs as a fissure filling in inertinite.

Inertinite group. In the coals of the Chipaque Formation, both fusinite and semifusinite degraded are observed. Funginite is also identified as colonies or crushed. The inertodetrinite is present in layers parallel to the stratification (M103), it has been derived from fusinita, semifusinita and funginite crushed and embedded with mineral matter (siderite, clay). Micrinite has also been observed in these coals, replacing liptinite macerals as bands or in the form of elongated lenses, and the macrinite is characterized by irregular and sub-rounded forms.

Mineral matter. The mineral matter is sulfide (pyrite) in disseminated, euhedral, and framboidal form, carbonate, and clay that fill cracks and replace liptinitic macerals, mainly cutinite.

Micro-lithotypes. Bimaceral is the dominant group. The highest concentration is found in M106, with values between 61.23 and 70.99% vol, and an average value of 66.11% vol. Clarite as the dominant micro-lithotype shows the highest value of volume percentage in the M106 seam (34.53 to 63.49% vol), with an average value of 49.01% vol. After the bimaceral group the next dominating association is the trimaceral with the highest frequency between 27.27 and 59.24% vol, and an average value of 43.46% vol. In the M101 seam the vitrinertoliptite prevails with the highest frequency between 17.29 and 31.3% vol. The quantitative distribution of micro-lithotypes of the Chipaque Formation is shown in Table 3.

Palmichal Group

The quantitative distribution and photographs of macerals of the Palmichal Group are shown in Table 2 and Fig. 6. For this geological unit, the M151 seam presents average values for the three maceral groups (20-30%) with a prevalence of collotelinite, liptodetrinite and inertodetrinite. The mineral matter analysis shows clay, carbonates, and pyrite varying between 8.8 and 12.8%.

Vitrinite group. The vitrinite group occurred at a low concentration (34.46% vol). Telovitrinite is the dominant subgroup and collotelinite is the most abundant maceral. The average value of collotelinite is 15.03% vol, and it occurs in thick bands. Second in frequency is the gelovitrinite subgroup, with significant values of both corpogelinite and gelinite (8.09 to 3.46% vol). The corpogelinite is found as isolated bodies with oval and rounded shapes or filling empty spaces. The gelinite is observed homogeneous appearance, filling cavities without a defined structure, it reflects an advanced process of devolatilization. The collodetrinite is observed as a groundmass that binds other macerals, while the vitrodetrinite as small particles of vitrinite of variable shapes.

Liptinite group. The average content of the liptinite group is 28.28% vol. The most frequent macerals are liptodetrinite, cutinite, alginite, and resinite, in that order, and a low concentration of liptinite macerals are represented by sporinite, fluorinite, bituminite, and exudatinite. Liptodetrinite (6.23% vol) occurs as fragments of other liptinitic macerals. The cutinite (6.01% vol) is characterized by a well-preserved thin wall (tenuicutinite), with jagged edges, occasionally it can be deformed and is observed toward the borders of the collotelinite. The alginite (5.46% vol) appears as colonies of *Botryococcus* and shows spherical and ovoid shapes. The resinite occurs as irregular bodies or filling cavities, and its rounded edge is common. The sporinite is observed as microsporinite and megasporinite, with well-preserved individual bodies, in which the internal morphology is occasionally identified. Between the secondary macerals, the fluorinite is characterized by its intense yellow fluorescence, and is recognised as a product of devolatilization, frequently associated with cutinite. Another secondary maceral is the exudatinite (0.90% vol), mainly observed filling the cavities of fusinite, whereas the bituminite (0.49% vol) is observed as an amorphous groundmass involving other macerals.

Inertinite group. The inertinite group is present with concentrations between 35.09 and 41.27% vol, with an average of 37.81% vol. The inertodetrinite is the most abundant maceral of the group, with an average value of 12.83% vol it is observed as debris without a defined structure, characterized by small remnants of decomposed and fusinitized plants within the peat (Nowak and Nowak 1999) as the result of the macerals transport (Scott 2002). The semifusinite (8.39% vol) is observed without cell structure since it is not well preserved. Funginite (7.13% vol) occurs as bands parallel to the stratification or in agglomerated shapes. Secundary inertinitic macerals are represented by macrinite, which is distinguished as amorphous bodies of different sizes (1.95% vol), and micrinite (2.42% vol) in lenses.

Mineral matter. Clay, sulphides (pyrite) and carbonate are the minerals included in these coals, the average frequency value of the mineral matter is 11.13% vol. The minerals occur as groundmass, cracks, fissures, or cleat fillings.

Micro-lithotypes. The macerals have been observed as trimaceral form (53.92% vol). The vitrinertoliptite is the most abundant micro-lithotype recorded (24.36% vol.). The quantitative micro-lithotype distribution of the Palmichal Group is shown in Table 3.

Arcillas del Limbo Formation

The quantitative distribution and photographs of macerals of the Arcillas del Limbo Formation are shown in Table 2 and Fig. 7. The analyzed seams present huminite content not higher than 50% (28.8 – 46%), significant values of the liptinite group (27 – 35.2%) and low contents of inerts (up to 35.4%), whereas mineral matter is mainly clay with a low average value (1 – 8.4%), except for the M201-4 seam with 14.4%.

Huminite group. The main subgroup of the geological unit is the telohuminite, with the maceral dominance of the ulminite, in thick bands. The proportion of the detrohuminite and gelohuminite subgroups is significant, with the maceral prevalence of the densinite and corpohuminite, this is characterized by an oval or sub-rounded shape and is filling the empty spaces of a pre-existing matrix.

Liptinite group. The most common of the macerals are the sporinite, cutinite, and resinite while the liptodetrinite, fluorinite, bituminite, and exudatinite macerals are founded in lower concentrations. The average value of the maceral content of the liptinite group is 31.78% vol. Sporinite is identified as microsporinite and megasporinite with thick and jagged walled. Pollen is also found without defined shapes. The cutinite is observed with thick wall (crassicutinite) and dark brown colour. Resinite is characterized by oval and round shapes, and it is filling cell cavities or diffuse form. Low alginite concentration was observed in these coals. The secondary macerals occur as fluorinite, liptodetrinite, and bituminite (amorphous) in low proportions. Exudatinite is observed filling the cavities of the inertinite macerals preferably.

Inertinite group. The quantified concentration of inertinite is between 12.58 and 26.07% vol, with an average of 21.11% vol, with the inertodetrinite as the most abundant maceral, although fusinite, semifusinite, funginite, macrinite, and micrinite are also observed. Fusinite and semifusinite are preserved and in low proportions. The maceral funginite has moderate values, not higher than 5.59% vol. They are observed as round and oval bodies in groups of single cells, represented by teleutospores (fungal spores). Its low percentage may indicate low bacterial activity. Micrinite is present as lenses or thin bands and it can be associated with mineral matter. Fusinite, semifusinite and funginite detritus can be distinguished in the inertodetrinite. It is the most abundant maceral, with an average value of 9.58% vol in M202.

Mineral matter. The volume of mineral matter is observed mostly in the form of clay, pyrite, and carbonate, with an average frequency of 5.5% vol. It appears as groundmass, fissures, and cleat fillings.

Micro-lithotypes. Macerals are associate to the bimaceral form, with frequency values ranging between 36.01 and 64.52% vol, with an average of 47.04% vol, being clarite the predominant micro-lithotype (25.69 and 53.33% vol). The average value of the trimaceral form is 41.64% vol, and the prevailing micro-lithotype is vitrinertoliptite (14.34-16.27% vol), with an average of 15.31% vol, which is in concordance with the high percentage of liptinite in the seams. The quantitative distribution of micro-lithotypes of the Arcillas del Limbo Formation is shown in Table 3.

San Fernando Formation

The quantitative distribution and photographs of macerals of the San Fernando Formation are shown in Table 2 and Fig. 8. The huminite concentration in the San Fernando Formation is greater than 50% vol (58.56 to 80.61% vol), among the three macerals groups this is the most abundant. The content of the liptinite is 16 to 25.30% vol, and the concentration of inertinite is the lower (3.29 to 18.51% vol in the M304 seam). The mineral matter is not greater than 14.4% vol in the M303 seam and is represented by disseminated pyrite, clay, and occasionally carbonate.

Huminite group. This group is represented by ulminite (22.20 to 30.39% vol) and densinite (3.50 to 24.43% vol), except in the M303 seam, in which the percentage of attrinite is considerable (50.70% vol). The ulminite is observed in homogenous bands of variable thickness, with cracks and fractures filled with mineral matter, clay mainly. Textinite is identified by its well-preserved cellular structure. The lumens are observed as oval bodies and filled of gelohuminite and mineral matter mainly. The attrinite shows particle sizes <10 microns, with clay filling of the empty spaces between fragments.

Liptinite group. Sporinite and cutinite are frequently occurring in these coals, their concentration values reach up to 14.48% vol. These macerals are indicators of humid environments related to terrestrial plants (Diessel 1992). The resinite, as secondary maceral, presents its highest value in the M302 seam (7.59% vol). Sporinite is found as microsporinite, tenuisporinite (thin walled), in parallel arrangement to the bedding plane. Spores sacs (sporangium) are observed in the seams M302 and M303, which occurs in lenticular to elongated bodies, reddish-brown colour under reflected light observations, and yellow colour under fluorescent light, highlighting their internal morphology. The cutinite concentration is 8.24% vol. It is observed with as thin and thick walled; the edges are jagged and orange colour under fluorescent light. The alginate- *Botryococcus* type, is spherical or fan-shaped, grouped in colonies, it is of planktonic origin, united by a gelatinous tissue and typical of lacustrine environments during the deposition of the coals (Pickel et al. 2017). Fluorinite occurs as a sub-round shape in continuous layers or filled cavities, a bright yellow color is observed under fluorescent light. It comes from vegetable oils (Suárez and Crelling 2008). Resinite is present as an irregular globular shape and filling in empty spaces. It emits an intense yellow orange fluorescence, it has not an internal structure and comes from the excretion of cells from different parts of plants (Pickel et al. 2017).

Inertinite group. The highest concentration of inertinite has been observed in the M304 seam (19.51% vol), other coals have values not higher than 10.85% vol. The predominant maceral is semifusinite, with other secondary macerals as funginite, macrinite, and micrinite. Semifusinite is characterized by a well-preserved cellular structure. Funginite is identified by round or elongate bodies of sclerotia, whose spaces may be filled with exudatinite or mineral matter. Secondary macerals as macrinite and micrinite occur in low proportions, they are a consequence of an early stage of carbonization.

Mineral matter. The frequency value of the mineral matter is among 3.20 – 14.4 % vol. It includes mainly carbonate, sulphide (pyrite disseminated) and clay.

Micro-lithotypes. The macerals of the San Fernando Formation coals are grouped as monomaceral (M303 and M305) and bimaceral forms. The coals include high concentration of huminite and liptinite, thus the predominant microlitotypes are liptite (45.08% vol -M303 seam), and clarite (66.81% vol - M302). The quantitative distribution of micro-lithotypes of the San Fernando Formation is shown in Table 3.

Interpretation and discussion

The concept of coal facies is based on the restoration of the type of peat and flora that constitute the coal, from the evaluation of factors such as climate, level and chemistry of the water table, and the contributions of nutrients at the time of peat formation (Diessel, 1986; Calder et al., 1991; Suárez et al., 2012; Singh, 2016). Nevertheless, the use of petrographic data as the only source in facies analysis has been controversial by different authors, such as Crosdale (1992, 1993), Dehmer (1995), Wüst et al. (2001), Scott (2002), Moore and Shearer (2003), Diesel (2007), (Hower and Ruppert (2011), Sahay (2011), Sen (2016), Dai et al. (2020). Here, we summarize some of their main arguments that may discourage the use of analysis methods to develop facies models only with coal petrography:

- The models that are representing the coal range and the geological age in specific places makes problematic its use outside of the proper conditions.
- The models simplify the effects of humification on tissue preservation and destruction.
- The authors mention that model do not consider some changes in petrographic composition related to floral evolution, or range and compaction increase.
- Lack of distinction between the different inertinite maceral in some models.
- They refer studies on modern peat that do not support any of the inferred relationships.

- They indicate that Gelification Index (GI) only takes gelation into account during turbification, but not during coalification.
- Disagreement in the use of geochemical gelification to determine the depositional environments and the Tissue Preservation Index (TPI) to interpret tree density, since there are many trees that are not woody and woody plants that are not trees.
- Besides, the different gelation processes between angiosperms and gymnosperms are not considered in the models.

The present study shows the facies and the coal deposit environment, obtained from the petrographic indices and models, without forgetting the factors mentioned above. We argue that the models apply the fundamentals related to the active processes at the time of peat formation, the appearance, and the nature of the maceral groups and show coherence with other disciplines regarding the depositional environment analysis.

Three facies of coal were identified in the studied seams, as indicators of the active process at the time of peat formation -*preservation, degradation, or fusinitization* (Rimmer et al. 2000). Table 4.

The Facies A represents the San Fernando Formation, it is characterized by low content of Group III macerals as the fusinite, semifusinite, macrinite e inertodetrinite. A high concentration of Group II macerals reveals the degradation of the organic matter. Whereas a moderate content of the Group I macerals (<50%) are indicators of preservation and derived from woody tissue composed of cellulose and lignin (Suarez and Crelling, 2008), these are also characteristic of an environment of low humidity and low pH in forest peatlands (Diessel, 1992). The

Facies B has been observed in Arcillas del Limbo Formation, the Palmichal Group, and the Chipaque Formation, that contain Group II macerals, indicating the degradation of the plant and includes remnants of the macerals of the vitrinite group (detrovitrinite, corpogelinite), macerals of liptinite (sporinite, resinite, cutinite, exudatinite, liptodetrinite), and micrinite (Rimmer et al. 2000). The facies C has been identified in Chipaque Formation (M106-2, M104-2, M104-3, and M101-5), the seams contain the highest percentages of macerals of the vitrinite group (Group I), especially collotelinite as indicators to better preservation of the organic matter, additionally, the seams have a low liptinite concentration (Fig. 9).

The Mukhopadhyay (1986) model is defined by the maceral composition of the coals. The seams of the Chipaque and Palmichal formations are characterized by the content of vitrinite and liptinite of terrestrial origin (vertex A and ¨D zone¨ of the triangle, Fig. 10), which suggests that the peat was formed in a forest swamp in oxic-to-anoxic conditions, with good tissue preservation. Meanwhile, the Arcillas del Limbo Formation indicates greater oxidation, and the San Fernando Formation shows an increase of anoxic and bacterial activity (Fig. 10).

The ternary diagram by Singh and Singh (1996) suggests the formation of the coals in a moor with an alternation between oxic and anoxic conditions (Fig. 11).

The liptinitic macerals of the coals plot in the triangular model proposed by Singh and Singh (2000), which indicates that the environment of coal deposition is controlled by the water depth conditions of the peat swamp. From this, the deposit of the Piedemonte Llanero coals is assumed in an open-water marsh dominated by reed plants such as the tall vegetation, like the grass of humid zones (Fig. 12).

The micro-lithotype composition of the Piedemonte Llanero coals is plotted in the ternary diagram proposed and improved by Singh et al. (2013), whose vertices indicate the micro-lithotypes derived from a maceral group. According to it, the development of forest to reed swamps facies in the Chipaque Formation coals are identified, whereas for the Palmichal Group the diagram suggests facies of forest swamp with sudden eustatic changes. In the Arcillas del Limbo Formation, forest to reed swamps facies are interpreted, while the San Fernando Formation is related to open-water facies with the predominance of subaquatic plants (Fig. 13).

Two parameters formulated by Diessel (1982, 1985, 1986, 1992), denominated Gelification Index (GI) and Tissue Preservation Index (TPI), have been defined for the analysis of the level water that covers the peat. These indices can be used to deduce the rates of accumulation and subsidence of the basin (Singh and Singh, 2013). The GI also determines the degree of persistence of wet or dry conditions (Diessel 1992). TPI is the measure that corresponds to the ratio between preserved and not preserves macerals, the index quantifies the degree of humification suffered by the precursor macerals of the organic matter and the proportion of woody matter that contributes to the total groundmass of the peat (Singh and Singh 2000; Singh 2016). According to the maceral composition, four zones are clearly defined: (i) *terrestrial*, consisting of relatively dry, arboreal peat, and located on the water table; (ii) *telmatic*, where the level of the water that covers the peat is controlled by variations in the level of the water table or sea level; (iii) *limno-telmatic*, which is a transition zone between telmatic and underwater environments; and (iv) *limnic*, which corresponds to underwater environments (Diessel, 1992). The GI and TPI are calculated as under:

$$GI = \text{Vitrinite} + \text{Macrinite} / \text{Semifusinite} + \text{Fusinite} + \text{Inertodetrinite}$$

$$TPI = \frac{Telinite + Collotelinite + Semifusinite + Fusinite}{Collodetrinite + Macrinite + Inertodetrinite}$$

The TPI ratio of the analyzed coals ranges from 1 to 5, and GI ratio ranges from 1 to 9, even though the San Fernando Formation shows the higher values between 10 and 100. The plot of the total amount of the Piedemonte Llanero coals in the diagram of Diessel (1986) suggests that they were originated in limnic to limnotelmatic conditions, except the San Fernando formation that shows a transition to telmatic conditions in the seams coal (Fig. 14).

To determine the factors that control the peatlands, Calder et al. (1991) introduced the GWI and VI indices. The GWI index deals with the ratio of the strongly gelled tissue and poorly gelled tissue, depending on the water supply and the pH. The VI index corresponds to the contrast of macerals of forest affinity related to those of marginal herbaceous and aquatic affinity. The table 5 consolidates the classification of the peats.

$$GWI = \frac{Gelinite + corpogelinite + clay\ mineral + quartz + vitrodetrinite}{telinite + collotelinite + collodetrinite}$$

$$VI = \frac{telinite + collotelinite + semifusinite + fusinite + suberinite + resinite}{collodetrinite + inertodetrinite + alginite + liptodetrinite + sporinite + cutinite}$$

The VI ratio of the analyzed coals ranges from 0.3 to 4, while the GWI ratio ranges from 0.1 to 3. The plot of Calder et al. (1991) suggests the origin of the coals in the transition area of bog to swamp environments. The low GWI would indicate that the mires were rain-fed without the influence of the groundwater level (Calder et al. 1991). The value VI <3 indicates the domain of herbaceous vegetation (Suárez et al. 2012). The Piedemonte Llanero coals are located along the transition zone of ombrotrophic to mesotrophic peatlands, it is also observed an interdigitation to reotrophic conditions that define transitional environments with flood periods, where the water table is located over the peat surface (Moore 1987) (Fig. 15).

Based on the integration of the micro-lithotypes (inertite, durite), the ternary diagram of Smyth (1979, 1984) reflects the oxidation degree of the organic matter prior to the organic matter burial and the vitrinite content in order to determine the environment of coal deposition.

The Chipaque Formation coals were deposited in a micro-tidal environment dominated by waves in shallow bays and coastal marshes of an estuarine system, where Guerrero and Sarmiento (1996) assume a partially closed lagoon as a result of the sea level increase. We also found that these coals are related to low oxidation conditions as it is indicated by the low percentages of durite and inertite micro-lithotypes, and significant contents of vitrite and clarite micro-lithotypes.

The Palmichal Group coals were deposited in shallow bays, swamps, and coastal lagoons of an estuarine system (Lower Guaduas Formation in Guerrero and Sarmiento 1996 and Ulloa 1976), possibly along an open lagoon with the active tide and wave processes that are reflected in the moderate concentration of the vitrinitic, intertinitic and liptinitic micro-lithotypes. The Arcillas del Limbo Formation coals show the greatest oxidation, so they may have been deposited in an estuarine and coastal plain environment (Ulloa 1976 and Lower Socha Formation in Guerrero and Sarmiento 1996), possibly along an estuarine lagoon with an increase in fluvial influence. The content of intermediate micro-lithotypes is low, while the proportion of micro-lithotypes of monomaceral and bimaceral character is considerable, especially for the durite and inertite by the important concentration of inertinitic macerals. The inertodetrinite indicates high oxidation and desiccation of the peat, caused by intermittent descent in the local water table, and it is the last organic residue before the complete mineralization of the plant matter (Diessel 1992; Nowak and Nowak 1999). Cutinite´s thick walls protect against dehydration, indicating dry environments (Diessel 1992). The San Fernando Formation coals reflect a higher concentration of vitrite and clarite as a product of wooded peatlands and low oxidation, the reducing chemical conditions of these deposits favored the preservation of plant tissues and they may have been deposited in a lacustrine-coastal plain environment (Dueñas and Van der Hammen 2007; Bayona et al. 2007, 2008; Parra et al. 2008; Pardo and Jaramillo 2014 and Caballero et al. 2020) (Fig. 16).

The preservation processes of peat, or the estuarine environment that allowed the development of the analyzed coals, follow the definition of estuary given by Dalrymple et al. (1992). According to these authors, the term is supported in sedimentological criteria and defined as "the portion towards the sea of a flooded river system that receives sediment from river and marine source areas and contains sedimentary facies influenced by tides, waves and river processes."

The results obtained from the coal facies suggest the evolution from an estuarine system to deltaic (lacustrine), since the microlithotypes composition correlates with different dominant processes, between tidal, waves and fluvial environments, which is also supported in sedimentology, stratigraphy and palynology studies in the Eastern Cordillera (van der Hammen 1958; Ulloa and Rodríguez 1976b; Guerrero and Sarmiento 1996; Dueñas and van der Hammen 2007; Bayona et al. 2007, 2008; Parra et al. 2008; Pardo and Jaramillo 2014 and Caballero et al. 2020).

The coal facies exhibit an ombrotrophic to mesotrophic and rain-fed peatlands (GWI <0.5). The GI>1 indicates a limnic to limnotelmatic environment up to the Paleocene, and telmatic in the San Fernando Formation (late Eocene - Middle Miocene) based on Calder et al. (1991) and Diessel (1986) models. The macerals and their associations indicate that the organic matter was expose to variations in the water table with oxic and anoxic conditions, increasing oxidation in the Paleocene and bacterial activity in the Middle Miocene according to sample plotting in the Singh and Singh (1996) and Mukhopadhyay (1986) models. The maceral content and its characteristics are related to the vegetation type and the water table relationship. The vegetation type according to Singh and Singh (2000), Singh et al. (2013), Calder et al. (1991), and Diessel (1986) models, indicate transition from the forest swamps to reed swamps with herbaceous vegetation (VI <3) in the Chipaque, Palmichal Group and Arcillas del Limbo formations, while subaquatic plants domain in the San Fernando Formation (high percentage of Ulminite, low Gelohuminite, thin-walled microsporinite, low content of inerts, mono and bimaceral microlithotypes- Vitrite - Liptite- Clarite). The domain of angiosperm in Late Cretaceous (Guerrero and Sarmiento, 1996) and Lower Eocene formations evolve to Pteridofitas and mangrove vegetation for the Eocene- Oligocene formations (Pardo and Jaramillo, 2014).

Conclusions

Coal petrology is a key technique to collect indicators elements about the formation conditions, as well as the type of the organic matter and its deposit environment in the coal geological units. Together with other disciplines as stratigraphy, sedimentology, and palynology, coal petrology provides data and convincing interpretations of the depositional scenario. The use of the petrographic indices and ternary models is assertive support to the reconstruction of the peat environment, such as the active processes at the formation moment, type of organic matter, precursor material, water level, and permanence of the humid or dry conditions. The thesis of determining deposits conditions from the micro-lithotypes analysis is proper since the maceral associations are indicators of the conditions that could be varied in transitional and restricted depositional environments.

The studied coals studied along the Piedemonte Llanero correspond to 15 seams of the Upper Cretaceous to Miocene geological units, located in the regional structures of the Boquerón, Recetor, Porvenir, Nazareth and Río Amarillo synclines, all of them with SW-NE trend and bordered by the Tesalia and Guaicáramo fault systems to the west and east, respectively. Coal petrology analysis shows a gradual increase in the range according to the depth, from the C sub-bituminous in the San Fernando Formation up to the most mature high-volatile C bituminous of the Cenomanian Chipaque Formation. Petrology also reveals the associated peats to the coals, varying from ombrotrophic to mesotrophic types, well extended, with arboreal and herbaceous affinity (rich in lignin and cellulose) and good tissues preservation. Related to the depositional conditions of the coals, the study manifests a restricted environment to the base of the sedimentary sequence in an estuarine system that evolves to a lacustrine environment of the deltaic system to the top.

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FIGURES CAPTIONS

Fig. 1 Geological map and localization of coal seams studied

Fig 2 Summary of nomenclature, palynology, and depositional environment for the coal of the Piedemonte Llanero

Fig. 3 Type of kerogen coal classification and maceral composition of Piedemonte Llanero coals plotted on a Van Krevelen diagram (1961 1993). Modified from Cornelius 1978 in Suárez and Crelling 2008

Fig. 4 Maceral composition of Piedemonte Llanero coals of Eastern Cordillera Colombia

Fig. 5 Microphotographs of macerals of the Chipaque formation. A) Chlorophyllinite in M101. B) Fusinite whose empty spaces are filled by exudatinite in M101. C) Inertodetrinite in M101-2. D) Thick-walled cutinite in M102. E) Inertodetrinite embedded in siderite in M102. F) Semifusinite degradaded in M103. G) Liptinite under fluorescence in M104. H) Banding of collotelinite with syngeneic framboidal pyrite in M104. I) Thick-walled megaspore in M104. J) Bands of collotelinite micrinite replacing liptinite in M105. K) Devolatilized vitrinite in M106. L) Collodetrinite containing inertodetrinite and clay mineral matter in M107. M) Bituminite in M101. N) Diagenetic pyrite in M101. O) Textinite in M103. Vitrinite (Vi) Textinite (Tx) Collotelinite (Ct) Collodetrinite (Cd) Fusinite (F) Semifusinite (Sf) Micrinite (Mi) Inertodetrinite (Id) Sporinite sporangium (Sp) Cutinite (Cu) Resinite (Re) Bituminite (Bi) Chlorophyllinite (Cl) Exudatinite (Ex) Clay mineral matter (Cy) Pyrite matter mineral (Py)

Fig. 5 (continued). Microphotographs of macerals of the Chipaque formation. P) Inertodetrinite in M102. Q) Resinite in M101. R) Original tissues preserved in M104. S) Mosaic of macerals in M103. T) Sporangium in M101. Vitrinite (Vi) Textinite (Tx) Collotelinite (Ct) Collodetrinite (Cd) Fusinite (F) Semifusinite (Sf) Micrinite (Mi) Inertodetrinite (Id) Sporinite sporangium (Sp) Cutinite (Cu) Resinite (Re) Bituminite (Bi) Chlorophyllinite (Cl) Exudatinite (Ex) Clay mineral matter (Cy) Pyrite matter mineral (Py)

Fig. 6 Microphotographs of Palmichal group macerals. A) Fluorinite in M151-3. B) Inertinite-semifusinite macrinite and fusinite whose empty spaces are filled by exudatinite in M151-2. C) Collotelinite bands limited by cutinite and with patches of inertodetrinite. Pyrite epigenetic-early diagenesis in M151-3. D) Vitrinite in the process of devolatilization and gelation (corpogelinite) in M151-2. E) Gelinite corpogelinite and thin-walled cutinite (tenuicutinite) in M151-2. F) Corpogelinite and resinite filling voids in M151-2. G) Thick-walled megaspore under fluorescence in M151-2. H) Alginite and *Botryococcus* colonies in M151-3. I) Inertinite in collodetrinite ground mass thin-walled cutinite deformed in M151-3. J) Epigenetic pyrite and fusinite in M151-3. K) Liptinite under fluorescence in M151-3. L) Inertodetrinite and vitrodetrinite in M151-3. Vitrinite (V) Collotelinite (Ct) Collodetrinite (Cd) Vitrodetrinite (Vd) Corpogelinite (Co) Gelinite (Ge) Fusinite (F) Semifusinite (Sf) Macrinite (Ma) Inertodetrinite (Id) Sporinite (Sp) Cutinite (Cu) Resinite (Re) Alginite (Al) Fluorinite (Fl) Exudatinite (Ex) Pyrite mineral matter (Py)

Fig. 7 Microphotographs of macerals of the Arcillas del Limbo formation. A) Funginite (teleutospore) and sporinite in M202-2. B) Macerals of the huminite group with sporinite inertodetrinite and resinite in M201-1. C) Sporinite and resinite in M201-1 with fluorescence. D) Ulminite band inertodetrinite and teleutospore sacs (funginite) in M201-1. E) Gelinite corpohuminite and inertodetrinite in M201-2. F) Fusinite and agglomeration of funginite in M201-2. G) Ulminite bands densinite that surrounds inertodetrinite macrinite sporinite and corpohuminite in M201-4. It is fractured. H) Textinite in M201-4. I) Corpohuminite resinite and semifusinite in M201-1. J) Micrinite and ulminite bands in M201-3. K-L) Reflected and fluorescent light. Ulminite with corpohuminite teleutospore (funginite) cutinite and resinite in M201-3. Textinite (Tx) Ulminite (Ul) Densinite (De) Corpohuminite (Ch) Gelinite (Ge) Fusinite (F) Semifusinite (Sf) Funginite (Fu) Macrinite (Ma) Micrinite (Mi) Inertodetrinite (Id) Sporinite (Sp) Cutinite (Cu) Resinite (Re)

Fig. 8 Microphotographs of macerals of the San Fernando Formation. A) Ulminite (Ul) bands containing microspores-tenuispores inertodetrinite and framboidal pyrite in M305. B) Textinite that envelops mineral matter (pyrite clay) and inertinite such as funginite and inertodetrinite in M305. C) Liptinites with fluorescence (resinite and sporinite) inM304 D) Thin wall cutinite (fluorescence) well preserved in M304. E) Ulminite bands interrupted by fractures deformed cutinite and sclerotinite- funginite equally affected in M303. F) *Botryococcus* algae colonies of planktonic origin in M302. G) Microspore sporangium in M302 and M301. H) Attrinite in M301. I) Fluorinite under fluorescence in M301. J) Microsporinite (under fluorescent light) in a ground mass of densinite in M301. K) Textinite in M301. L) Corpohuminite and ulminite bands in M301. Textinite (Tx) Ulminite (Ul) Densinite (De) Attrinite (At) Corpohuminite (Ch) Funginite (Fu) Inertodetrinite (Id) Sporinite- sporangium (Sp) Cutinite (Cu) Resinite (Re) Alginite (Al) Fluorinite (Fl) Pyrite mineral matter (Py)

Fig. 9 Ternary diagram illustrating genetic facies of Piedemonte Llanero coals. Modified from Rimmer et al 2000

Fig. 10 Ternary diagram corresponding to the model of facies of coals and suggested peat forming environment (modified from Mukhopadhyay 1986). A) Huminite + terrestrial liptinite. B) Humodetrinite + liptodetrinite + gelinite. C) Inertinite. D) Forest swamp halfway between oxic and anoxic with good tissue preservation. E) Reed marsh with increased maceration and bacterial activity and an anoxic increase. F) Dry condition (oxidant)

Fig. 11 Ternary diagrams suggested by Singh and Singh 1996 for coal facies of the Piedemonte Llanero. D. Alternate oxic and anoxic moor. E. Oxic (dry) moor with sudden high flooding. F. Wet moor with intermittent moderate to high flooding

Fig. 12 Ternary diagrams for facies models controlled by water depth conditions of peat swamp (modified from Singh and Singh 2000) in the Piedemonte Llanero coals. A) Area for the development of algal bloom and the zone of anaerobic bacterial activity. B) Open-water swamp dominated by reed plants. C) Transition between forest swamp and reed swamp

Fig. 13 Ternary diagrams for facies of coal of the Piedemonte Llanero. (modified from Singh et al. 2013). A = liptite + clarite (L) + vitrinertoliptite + durite (L) B = vitrite + clarite (V) + vitrinertite (V) + duroclarite C = inertite + durite (I) + clarodurite + vitrinertite (I). The types of facies delineated are CF1 deep-water facies characterized by organic mud rich in liptinites. CF2 open water with dominance of subaquatic plants. CF3 forest swamp with sudden eustatic changes. CF4 forest to reed swamp. CF5 forest swamp. CF6 moss swamp with extremely dry conditions

Fig. 14 Coal facies obtained from GI index and TPI of Piedemonte Llanero coals in relation to depositional setting and type of mire (after Diessel 1986 and modified by Kalkreuth et al. 1991). A. Vitrinite>Inertinite Degraded vitrinite> structured vitrinite B. Vitrinite>Inertinite Structured vitrinite> Degraded vitrinite C. Inertinite>Vitrinite Inertodetrinite> semifusinite+fusinite D. Inertinite>Vitrinite Semifusinite+fusinite> Inertodetrinite

Fig. 15 Mire paleoenvironmental diagram of Piedemonte Llanero coals in reference to Grondwater Index- GWI vs Vegetation Index- VI (modified after Calder et al 1991)

Fig. 16 Microlithotypes ternary diagram of the Piedemonte Llanero coals showing the evolution of an estuarine to deltaic system (adapted from Smyth 1979 1984)

TABLES CAPTIONS

Table 1.

Physicochemical results (wt %) and reflectance measurement (%) of Piedemonte Llanero coals in the Eastern Cordillera of Colombia

Table 2

Frequency distribution of maceral composition in vol. % in the Eastern Cordillera of Colombia

Table 3

Frequency distribution of microlithotypes composition in vol. % in the Eastern Cordillera of Colombia

Table 4

Distribution of macerals to genetic groups I II III. From Rimmer et al (2000)

Table 5

Classification of peatlands. From Diessel (1992) in Thomas (2012)