



Article Validating Structural Styles in the Flysch Basin Northern Rif (Morocco) by Means of Thermal Modeling

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Abstract: Vitrinite reflectance and a micro-Raman spectroscopy parameters data set have been acquired on dispersed organic matter of the Maghrebian flysch basin and the Tangiers unit across a NE-SW section in the north-western Rif belt (North Morocco). Thermal maturity shows increasing values from the hinterland to the external unit (from NE to SW). Paleo-thermal indicators show that the internal flysch basin (i.e., the Mauretanian unit) is less mature than the external one, (i.e., the Massylian unit), with R_0 % and R_0 eq. Raman values ranging from 0.64% to 1.02% (from early mature to late mature stages of hydrocarbon generation). 1D thermal modeling estimates the overburden now totally eroded ranging from 3.1 km to 6.0 km, and has been used as constraint to reconstruct the complete thrust wedge geometry in Miocene times. The reconstructed geometry accounts for high shortening (about 63%) due to the development of an antiformal stack in the frontal part of the wedge made up by the flysch succession. This stacking is interpreted as a consequence of the western translation of the Alboran Domain in the core of the Betic-Rif orogenic system.

Keywords: Rif belt; Maghrebian Flysch basin; vitrinite reflectance; micro-Raman spectroscopy; thermal modeling; thrust wedge

1. Introduction

Fold-and-thrust belts are structures through which former passive margin sedimentary covers and shallow basement rocks mainly shorten because of convergence. The rules of their evolution both across and along the strike are well-known [1–5]. Nevertheless, uncertainty exists in defining the structural style and shortening in the Central-Western Mediterranean region, where orogens develop with typical arcuate shapes [6,7] as a result of the long-lasting tectonic evolution in between the convergence of Eurasia and Africa. In particular, ranges of shortening can differ up to one order of magnitude [8], with important implications on energy resource potential evaluation in structurally complex areas [9].

The Rif belt (Figure 1) is one of the most complex areas in Western Mediterranean as it is extremely arcuate [10,11]. For many decades, the area complexity has brought the development of different and sometimes contrasting geodynamic models [12]. The most accepted models that explain the evolution

of the West-Mediterranean region are: (i) convective removal of the lithospheric mantle and orogenic collapse [13], (ii) a single subduction plane with multiple roll-backs [14], (iii) the Slab retreat model [15], and more recently, (iv) the double subduction model [16].

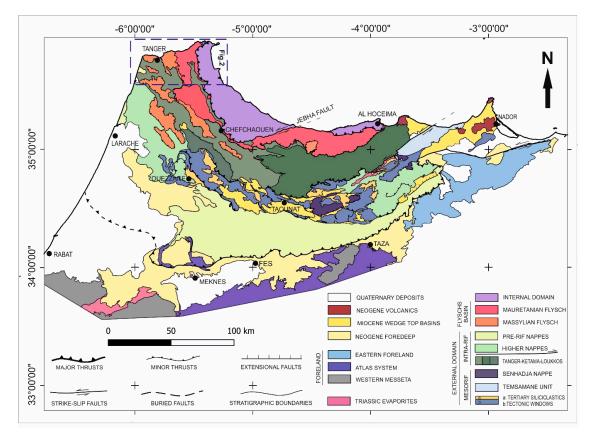


Figure 1. Structural sketch map of the Rif belt in northern Morocco (modified after [17–19]). Black dashed rectangle shows the location of the studied area, shown in Figure 2.

Such differences are the result of the lack or poor-quality subsurface data and/or scarce information related to the eroded portions of the belts themselves [20–23]. There is a lack of information bias on the reconstructions, mainly of the internal portions of the fold-and-thrust belts, which are characterized by intense deformation and detachments [11,24]. In such cases, structural tools, such as restoration and structural balancing techniques, allow multiple viable interpretations [25,26]. Moreover, in order to reduce the number of structural solutions, the estimation of the eroded portion of a thrust wedge through time is pivotal. To reach this aim, a useful tool is the integration of thermal modeling of sedimentary successions with section drawing [9,22,27].

In this paper, along a NE-SW regional transect crossing the Maghrebian flysch basin and the Tangiers unit (Intrarif Domain), eight 1D thermal models have been established to reconstruct the eroded portion of the shortened section to reduce uncertainties for section building. Thermal models were calibrated by the means of two different paleothermal indicators: vitrinite reflectance (R_0 %) and R_0 equivalent from Raman spectroscopy derived from analyses on dispersed organic matter.

2. Geological Setting

The Gibraltar arc represents the western edge of the West-Mediterranean Alpine systems. Its formation and arcuate shape [7,28,29] developed as a consequence of the westward translation of the Alboran Domain in the general framework of the Africa-Eurasia collision [30–33].

The Rif belt (Figure 1) represents the southern limb of the Gibraltar Arc. It is also a part of the Maghrebides (Tell-Rif) orogenic system [34], resulting from the progressive closure of the Maghrebian

Tethys and the final docking of the Alboran domain onto the African margin during the Late Burdigalian age [10,18,35].

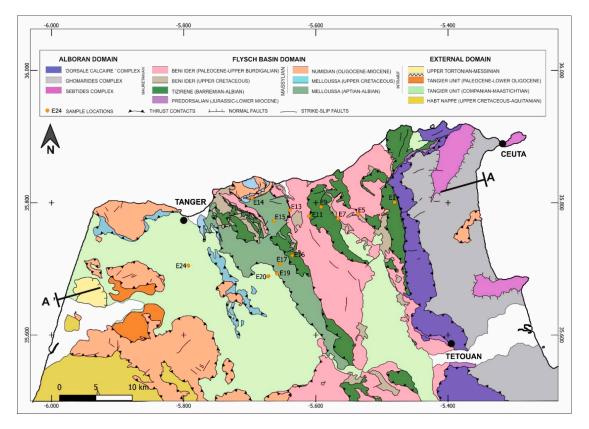


Figure 2. Geological map of the northwestern corner of the Rif belt with sample locations, compiled from the Geological maps of the Rif at 1/50000 published by the Geological Survey of Morocco [36–40]. A-A' refers to the studied transect (see Figure 4).

The Rif belt was built mainly during Miocene times by the interaction between the Alboran domain, the Maghrebian flysch basin, and the External domain [41,42]) toward the southwest and south (Figures 1 and 2).

The innermost Alboran domain [43,44] is derived from the northern paleo-margin of the Maghrebian Tethys and consists of the Sebtides Complex [44], which corresponds to continental terranes affected by Variscan and Alpine metamorphism [45,46]; the Ghomarides complex [47,48], which is made of Paleozoic terranes affected by Variscan metamorphism [48–50], except for the lowest part, which is also affected by a localized Alpine metamorphism [51]; and the "Dorsale Calcaire," which corresponds to Triassic-Lower Jurassic carbonates and Cenozoic siliciclastic deposits [52,53].

The Maghrebian flysch basin [10,54,55] corresponds to the Meso-Cenozoic deep marine southern branch of the Maghrebian Tethys, which was connecting the Alpine oceans with the central Atlantic from the Jurassic up to the Miocene [47,55–57]. The Maghrebian flysch basin is subdivided into internal Mauretanian and external Massylian Domains (Figure 3, [58–60]).

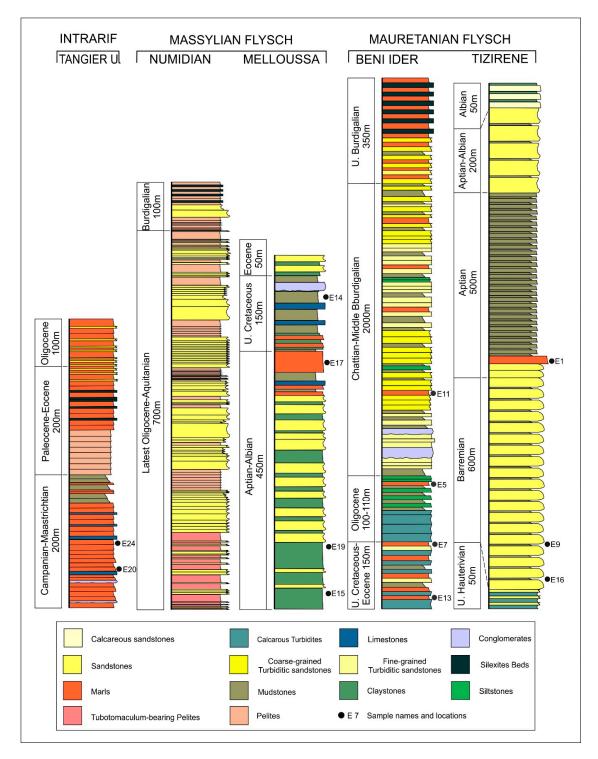


Figure 3. Simplified stratigraphic columns of the sampled units with sample names and positions. The stratigraphic columns are modified and/or redrawn after [18,57,61,62]. The columns are not to scale.

The transition between the two sub-domains is differentiated in the Oligocene-Lower Miocene sediments, where the Mauretanian Beni Ider turbidites evolve laterally to the Massylian Numidian sandstones through mixed successions [63–65]. During the orogenesis, the sedimentary cover of the Maghrebian flysch basin formed a series of thrust sheets (Figure 4) [65,66].

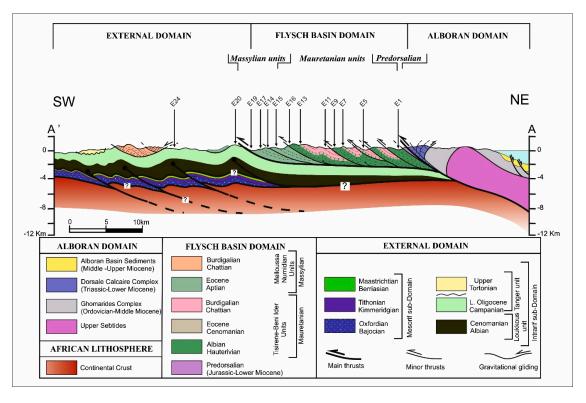


Figure 4. Structural cross-section (see Figure 2 for location) showing the overall architecture of the Maghrebian flysch basin and its relationship with the Internal and External domains based on [36–40] and on field observations. Sample names and position are indicated.

The external zones belongs to the North African paleomargin that was overthrust by the Internal and Flysch domains during the Cenozoic Alpine cycle [19,47,67]. The external Rif is subdivided into the Intrarif, which also organized into three main sub units, known as Ketama, Loukkos, and Tangiers units; the Mesorif; and the Prerif [17]. In front of the system, the Middle Atlas system represents the foreland of the Rif belt, where a Miocene foredeep basin developed. The latter is covered by the Prerif nappes and other more organized and far-travelled nappes (higher nappes) deriving from the Intrarif (Figure 1), namely the Aknoul, Tsoul, Habt, and Ouezzane Nappes [10,19,68–70].

3. Samples and Methods

3.1. Samples

Organic matter optical analysis and micro-Raman spectroscopy were performed on dispersed organic matter from thirteen samples carried out along a NE-SW transect (Figure 4). Three samples were collected from marls/mudstones levels of the Mauretanian Tizirene flysch, four samples from the marly levels of the Mauretanian Beni Ider flysch, four samples from the marl/claystone/mudstone levels of the Massylian Melloussa flysch, and two samples from the marls levels of the Intrarifain Tangier unit (Figures 2 and 3). All samples were collected far from major faults (e.g., >10 m), see (Figure 4), in order to avoid frictional heating and/or alteration effects [71].

3.2. Methods

3.2.1. Organic Matter Optical Analysis

Whole-rock samples were slightly crushed in an agate mortar, mounted in epoxy resin, and then polished according to standardized procedure [72]. Vitrinite reflectance (R_0 %) measurements were performed on randomly oriented organic grains with a Zeiss Axioplan microscope, using conventional

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micro-photometric methods under oil immersion (n = 1.518) in reflected monochromatic non-polarized light (λ = 546 nm). The calibration is based on three reflectance standards with a R_o% values of 0.427, 0.585, and 0.905 for Spinel, Sapphire, and Yttrium-Aluminum-Garnet, respectively. The population of the measured fragment varies between eight in samples with small amounts of organic matter and 40 in organic matter-rich ones. Measurements were performed on non-oxidized fragments, never smaller than 5 µm, only slightly fractured and/or altered [73].

3.2.2. Micro-Raman Spectroscopy on Organic Matter

The analysis of carbonaceous material by means of Raman spectroscopy has grown quickly in the last years. It has turned out to be a powerful geothermometer for metamorphic rocks [74–76], and recently also for coals and dispersed organic matter in diagenesis [76–86], even if a unique procedure has not still been defined [87].

In diagenesis, the first-order Raman spectra are composed by two main bands known as the D and G bands [88,89], and also other minor bands. The G band is linked to the in-plane vibration (E_{2g} symmetry) of carbon atoms in graphene in crystalline graphite at 1582 cm⁻¹ [88,90,91]. The D band at 1350 cm⁻¹ [92] becomes active in disordered graphite, and it has been interpreted as a result of a the double resonant Raman scattering process [90–92] or alternatively, it is able to rise from the ring breathing vibration in the graphite sub-units or polycyclic aromatic compounds [82,93–95] or from the aromatics with six rings or more [96]. The assignment of the other bands is still a matter of debate [85,94,96–99].

The best relationship between Raman parameters and thermal maturity in diagenesis is generally found for the full width at half maximum (FWHM) of the G and D bands, the position of the G and D band, the D/G area, and D/G intensity ratio (see [87] for a review).

Micro-Raman spectroscopic analyses were performed on whole-rock powders using a Witec-Rise Micro-Raman spectrometer, calibrated against the 520.74 cm⁻¹ band of silica. Data were collected over the first order Raman spectrum (700–2300 cm⁻¹) [88], using a 600 grooves/mm grating and CCD detector. A green laser ($\lambda = 532$ nm) with a power of 75 mW was used as light source, while the optical filters adjusted the laser power at <0.4 mW. To reduce the fluorescence, the Raman backscattering was recorded after an integration time of 30 s for four repetitions [85]; then between 10 and 30 measurements per sample were performed in order to ensure reproducibility.

The first analytical process performed on the spectra was the removal of the fluorescence background interfering with the Raman spectra of highly disordered carbonaceous material [74,85]. A linear baseline subtraction has been applied at the limits of the D and G spectral regions, respectively [100]. The D band is centered at about 1350 cm⁻¹, and thus, the D band region has been defined between 1100 and 1470 cm⁻¹, while for the G centered at about 1600 cm⁻¹, we defined its region between 1450 and 1700 cm⁻¹.

Despite the fact that Raman spectroscopy has been widely used on dispersed organic matter and coals to assess thermal maturity, its application in the "oil window" is still a matter of debate, essentially due to the different curve-fitting methods in this temperature range [84,86]. In this work we adopted the automatic approach proposed by Schito and Corrado (2018) that requires a minimum data manipulation. In this method, D and G bands are fitted separately, applying a one-band asymmetrical Gaussian deconvolution for each spectral region, with an asymmetry of 65% and 60% for the D and G bands, respectively. We determined position, intensity, width, and integrated area of the D and G bands. The applicability range is between 0.3 and 1.5 R_0 % [100].

The Raman spectral parameters are used to calculate the Ro equivalent % (R_o% eq.) through a parametric equation resulting from a multi-linear regression based on a correlation between Raman parameters and vitrinite reflectance (R_o%) [100].

3.2.3. Thermal Modeling

Thermal history and burial of the northwestern side of the Maghrebian flyschs basin were performed using the Basin Mod 2-D software package by Platte River (2020). Assumptions used for the modeling are: (1) decompaction of the burial curves according to Sclater and Christie [101]; (2) thrusting duration is considered instantaneous [102]; (3) a sediment-water-interface temperature of 12 °C and a surface temperature of 20 °C are assumed; (4) burial and thermal models were constrained by organic thermal indicators (vitrinite reflectance R_0 % and R_0 % eq. [100]), and are carried out using the LLNL Easy%Ro method based on the kinetic model of vitrinite maturation of [103] and [104]; (5) heat flow is fixed at 55 mW/m² [105,106]; (6) thicknesses, lithology, and ages of sediments are from [18,57,61,62], and have been used to constrain the pseudo-wells perpendicularly to the main stratification while respecting the samples positions; (7) finally, the sea-level variations are neglected, as the thermal evolution is mainly controlled by sediments thickness [107].

4. Results

4.1. Organic Matter Optical Analysis

Vitrinite reflectance values in the area vary between 0.64 and 1.02% (Table 1).

Table 1.	Organic matter	maturity	derived	from	vitrinite	reflectance	$(R_0\%)$	and	micro-Raman
spectroscopy (R ₀ % eq.) analyses.									

UNIT	SAMPLES	COORDINATES	AGES	R _o %	Sd. (±)	Nr. Fr	R _o % eq.	Sd. (±)
	E1	35.80055556; -5.480833333	Aptian	0.64	0.08	9	_	_
MAURETANIAN	E5	35.78333333; -5.535833333	Oligocene	0.92	0.08	16	_	_
	E7	35.775963; -5.566249111	Eocene	0.65	0.08	12	_	_
	E9	35.79361111; -5.592222222	Barremian	0.75	0.07	23	-	_
	E11	35.77958303; -5.610247222	Miocene	1.00	0.08	40	_	_
	E13	35.78833333; -5.639166667	Campanian	0.84	0.04	8	0.86	0.08
	E16	35.72027778; -5.635833333	Barremian	0.98	0.05	29	-	-
	E15	35.77277778; -5.663611111	Aptian	1.01	0.07	18	1.00	0.08
MASSYLIAN	E14	35.80676; -5.696666	U. Cretaceous	0.77	0.06	20	-	_
	E17	35.70777778; -5.654166667	Albian	1.00	0.05	28	0.98	0.07
	E19	35.69285255; -5.658765111	Aptian	0.82	0.06	33	0.90	0.10
INTRARIF	E20	35.68888889; -5.671944444	Campanian	1.02	0.06	30	1.06	0.06
	E24	35.705; -5.792777778	Campanian	0.87	0.09	24	0.88	0.10

Coordinates system: "WGS-84"; Sd. (±): standard deviation; Nr. Fr: number of fragments.

The Mauretanian Tizirene unit constitutes four thrust sheets (Figure 4). The three samples studied along the NE-SW transect (Figures 2 and 4) contain well-preserved fragments belonging to both the vitrinite and inertinite maceral groups. The R_0 % values increase from internal to external thrust sheets according to their depth and structural position, and range from 0.64% to 0.98% (Figures 3–5), indicating the early to middle stages of oil generation [108].

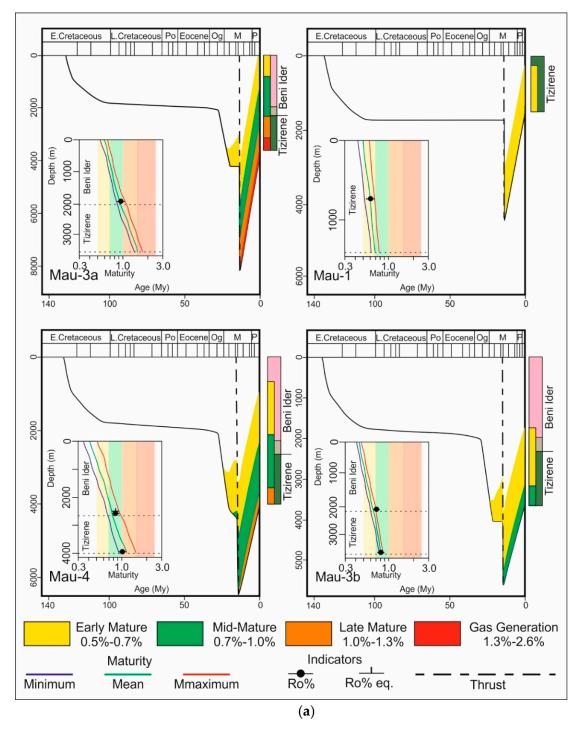


Figure 5. Cont.

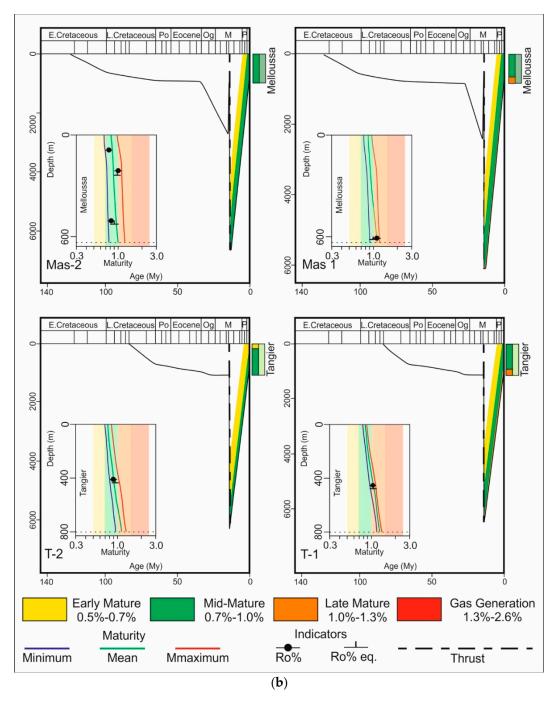


Figure 5. (a) Burial and thermal models of the Mauretanian thrust sheets showing maturity versus depth and hydrocarbon generations. Diagrams constrained by vitrinite reflectance R_0 % and R_0 % equivalent (calculated from Raman parameters). For detailed lithology, see Figure 3. Mau-(1, 3.a., 3. b., and 4): pseudo-wells belonging to each thrust sheet. Po—Paleocene, Og—Oligocene, M—Miocene, and P—Pliocene; (b). Burial and thermal models of the Massylian thrust sheets and Tangier unit showing maturity versus depth and hydrocarbon generations. Diagrams constrained by vitrinite reflectance R_0 % and R_0 % equivalent (calculated from Raman parameters). For detailed lithology, see Figure 3. Mas-1 and Mas-2: pseudo-wells belonging to the Massylian thrust sheets; T-1and T-2: pseudo-wells belonging to Tangier unit. Po—Paleocene, Og—Oligocene, M—Miocene, and P—Pliocene.

The Mauretanian Beni Ider unit is comprised of three thrust sheets (Figure 4) and contain well-preserved vitrinite and inertinite. The R_0 % values vary between 0.65% and 1.00% (Table 1), increasing from internal to external thrust sheets outlining maturities in the middle mature stages of

hydrocarbon generation. Vitrinite reflectance data from the Beni Ider unit show two different levels of thermal maturity along the transect, depending on the samples position with respect to the upper thrust. R_0 % values range from 0.65% to 0.84% when they are far from the tectonic contacts, and from 0.92% to 1.00% when the samples lie just below the upper thrust.

The Massylian flysch domain is represented by the Melloussa unit, composed of two thrust sheets (Figure 4). The four collected samples show the presence of both vitrinite and inertinite macerals groups. R_0 % values range between 0.77% and 1.01%, indicating the middle mature stage of hydrocarbon generation. The obtained values show a trend of increasing maturity with stratigraphic depth. Locally, shallow samples located at the footwall of the thrust separating the two thrust sheets show slightly higher values (Figures 3 and 4). Generally, the average values of R_0 % in the Melloussa unit are higher than in the Tizirene and Beni Ider units.

The Tangiers unit (Figures 2–4, and Table 1) provides R_0 % values of 1.02% at the footwall of the regional thrust located between the Maghrebian flysch basin domain and the Intrarif domain, and the 0.87% moving externally toward the West. The obtained values indicate the middle mature stage of hydrocarbon generation.

4.2. Micro-Raman Spectroscopy on Organic Matter

From the thirteen collected samples, six were suitable for micro-Raman spectroscopic analysis. However, all along the NE-SW section presented in Figure 4, the R_0 % equivalent values derived from the Raman spectra are highly consistent with the measured vitrinite reflectance R_0 % (Table 1). In the Mauretanian Beni Ider unit, the R_0 % equivalent value calculated from the Raman spectra is 0.86%. In the Massylian Mellousa unit, the R_0 % equivalent values are between 0.90 and 1.00%. In the Tangiers unit, the R_0 % equivalent values are 1.06% and 0.88%.

5. Discussion

Vitrinite reflectance is the most widespread thermal maturity indicator in the diagenetic realm [109], while Raman spectroscopy on organic matter has been widely debated, especially when related to thermal maturity assessment in diagenesis [77,85,87,110,111].

The use of different thermal maturity indicators in the thermal model is highly recommended since it has been demonstrated that vitrinite reflectance alone can suffer several pitfalls due to the scarcity or misidentification of maceral content, or reflectance retardation/suppression phenomena [112]. Due to scarcity or uncertainties derived from a small dimension of vitrinite fragments, we decided to couple optical analyses with Raman analyses on organic matter using the equation proposed by [100]. The good agreement between the two methods makes our results particularly reliable for thermal model calibration. Moreover, in order to provide a large spectrum of solutions, three different burial-thermal scenarios are presented providing minimum, mean, and maximum values. Among them, the mean values are used to discuss the overburden calculation for sake of simplicity. Two wells (Mau-3a and Mau-1) were calibrated with only one set of thermal maturity data, which is not usually considered enough to ensure model reliability. Nevertheless, assuming no significant variation in the heat flow regime, one point on the calibration curve can be sufficient to evaluate the magnitude of the tectonic burial.

In the Maghrebian flysch basin (Figures 1 and 2), the evolution of the Mauretanian succession started in Lower-Cretaceous times with the deposition of the Tizirene turbiditic flysch (Figure 3), and continues up to the Late-Burdigalian with the deposition of the Beni Ider siliciclastic sediments, which become gradually thicker toward the external portion of the Mauretanian basin [57]. During the Alpine orogeny, between the Late Burdigalian and Early Langhian, the Mauretanian and Massylian flysch were involved in the advancing of the orogenic wedge [66,113]. The compressive event affecting the Maghrebian flysch was expressed by four NE-SW thrusts sheets. Thermal models performed for the Mauretanian flysch basin show an increase of overburden moving from the most internal thrust

sheet to the most external one (Figure 5a), varying from 3.1 km in the more internal thrust (Mau-1), located immediately at the footwall of the 'Dorsale Calcaire', to 4.2 km in Mau-4 (Figures 4 and 6).

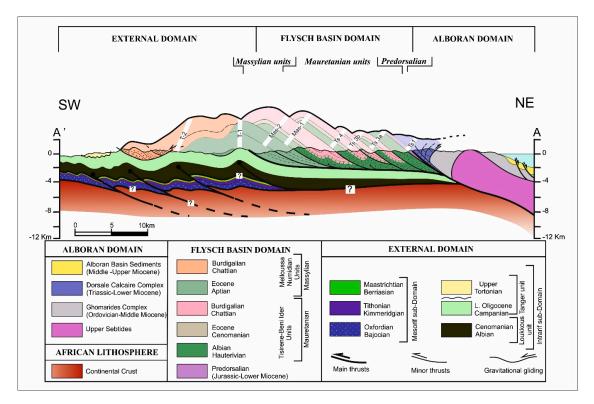


Figure 6. Structural cross sections showing the envelope of the paleo-wedge in Miocene times (black dashed bold line) and calculated tectonic loads. White bars represent the locations where thermal models were performed.

Moving toward the SW, the Massylian flysch basin was deformed into two thrust sheets (Mas-1 and Mas-2) (Figures 4 and 6) with a NE-SW direction. Two thermal models have been performed in the Massylian domain (Figure 5b). The evolution started from the Aptian-Albian times with the deposition of the Mellousa flysch, up to Late-Burdigalian marking the end of the thick Numidian sandstones deposition. In Langhian times, the Massylian successions suffered a tectonic burial of 5.4 km on Mas-1 and 6 km on Mas-2 (Figure 6). The emplacement of this tectonic load is synchronous with the activation of a regional flat within the Massylian domain between Mellousa and the Numidian. This detachment allowed the Numidian sandstone to glide in front of the Massylian domain onto the Tangiers unit. Accordingly, this succession experienced deep diagenetic conditions and a middle-mature stage of hydrocarbon generation.

More externally, two thermal models have been performed for the Tangiers unit, whose sedimentation started in Late Cretaceous time. During the progressive advancing of the wedge, the Tangiers unit underwent a tectonic burial at about -14 Ma ranging between 5.5 km (T-1) and 5 km (T-2) Figure 5b and Figure 6).

The discrimination between the sedimentary and tectonic contribution to the burial paths in the flysch basin is well constrained. First of all, the stratigraphy was defined by previous literature (Figure 3). Furthermore, possible younger sediments (unconformable Pliocene deposits [57]) crop up to the south of the study area, but are not recognized along the studied transect. Such evidence suggests that the overburden calculated from the models along the studied transect could be due to thrusting. These considerations allowed us to define the envelope of the thrust wedge by the projection of the calculated loads derived from thermal modeling onto the cross section (Figure 4), and to reconstruct the shape of the accretionary wedge in Miocene times before erosion (Figure 6).

The reconstructed wedge becomes higher toward its external part, shedding light on the lack of cylindricity along-strike in the fold and thrust belts, which is commonly expressed by the variation of structural styles and shortening rates [5]. Strike-slip faults are considered one of the main factors controlling the along-strike variations (e.g., [114]), and are usually debated in kinematic reconstructions. This feature should not be neglected in the Rif belt that is affected by the Jebha regional left-lateral strike-slip fault. The latter caused considerable differences in the geometry and internal deformation organization on both sides of the fault [115–118], and by consequence, controlled the shortening rates in the terranes cropping up north and south of this lineament (Figure 1). We suggest that the propagation of the orogenic wedge may be enhanced by this along-strike variation leading to the development of a thrust sheet stacking, which can be explained by the acquired tectonic loading and the 62.8% rate of shortening (Figure 6). Additionally, it is assumed that the Maghrebian flysch basin was deposited on an oceanic or thinned continental crust [10,54,56,57,65,113] bordered southward by the north African paleomargin, nowadays represented by the Intrarif. The latter may act as a bulge controlling the frontal propagation of the wedge, due to crustal thickness contrast. We consider this feature responsible for the development of the antiformal stacking in the most external side of the Maghrebian flysch basin, which is expressed by a tectonic loading reaching 6.0 km of thickness (Figure 6). In conclusion, we suggest that the shape of the reconstructed wedge during Miocene times (Figure 6) is, on one hand, the result of compressive tectonic events due to the Africa-Iberia convergence and the Alboran domain translation during the Alpine orogeny. This process is probably enhanced by the role of the Jebha fault and the north African paleomargin that acted as a bulge blocking the wedge propagation.

6. Conclusions

Organic thermal maturity within the flysch basin of the Rif belt shows a general increase moving from internal to external portions of the fold-and-thrust belt. Levels of thermal maturity for the outcropping successions range from early-mature to late-mature stages of hydrocarbon generation. Combining thermal modeling and the present-day geometry of the wedge, we can reconstruct the shape of the thrust wedge in Miocene times, which shows an increase in thickness moving toward the southwest (i.e., North African paleomargin) with tectonic loads ranging from 3.1 km to 6.0 km. The high shortening of 63% within the Maghrebian flysch basin highlighted the role played by the Jebha fault in the thrust wedge evolution as suggested by previous authors [115–118]. Both the thrust wedge propagation and the pre-orogenic geometry of the Maghrebian flysch basin and related margins are responsible for the formation of numerous thrust sheets and the development of an antiformal stacking at the front of the wedge, shortening the flysch basin domain.

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