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Original paper

Exploring metamorphic temperatures in the Kaczawa Metamorphic Complex using Raman spectroscopy of carbonaceous material

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Raman spectroscopy was used to measure the thermal maturity of organic matter in widespread low-grade Ordovician and Silurian metasedimentary rocks of the Kaczawa Metamorphic Complex. The suitability of different Raman-based geothermometer formulations for estimating peak metamorphic temperatures in the studied rocks was evaluated. Among the tested geothermometers, the formulation based on the full width at half maximum of the D1 band, and the calculations performed with the IFORS software yielded most consistent results, reproducing with sufficient accuracy and reliability the peak metamorphic temperatures obtained by other authors. Disruptions in the regional temperature distributions due to thermal overprint caused by local volcanic activity were identified by studying the contact aureole around a large body of the Wielisławka Rhyolite. It was found that the influence of contact metamorphism on the measured spectral parameters becomes insignificant at about 150 m away from the contact. The regional distribution of peak temperatures not affected by late thermal overprinting shows a clear pattern with the record of higher temperatures preserved in the southern part of the Kaczawa Metamorphic Complex. Depending on the tectonic unit, peak temperatures vary from 312 to 352 °C, with an average of about 331 °C. Temperatures calculated for the units in the northern Kaczawa Metamorphic Complex branch range from 278 to 301 °C, with an average of 289 °C. With estimated geothermal gradient of 15 °C/km for the peak thermal conditions, the difference of 40 °C indicates the southern branch was buried 2.5–3.0 km deeper than the northern branch.

Keywords: Kaczawa Metamorphic Complex, geothermometry, Raman spectroscopy, organic matter, low-grade metamorphism, Sudetes
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1. Introduction

The Raman spectroscopy of organic matter in metasedimentary rocks has recently become the most widely used geothermometer (Schito et al. 2023), with its principles described in several Raman reviews (e.g. Lünsdorf et al. 2014; Henry et al. 2019; Schito et al. 2023), detailed geothermometer development contributions (Beyssac et al. 2002; Rantitsch et al. 2004; Rahl et al. 2005; Aoya et al. 2010; Lünsdorf et al. 2017), or geothermometer application papers (e.g. Cheshire et al. 2017; Lupoi et al. 2017; Khatabi and Aghajanpour 2018; Beyssac et al. 2019). A number of parameters based on the relative proportions or absolute values of band height, width, area, position, or band separation obtained from Raman spectra are utilized for this purpose. These parameters are often calibrated to metamorphic temperatures (Beyssac et al. 2002; Rantitsch et al. 2004; Rahl et al. 2005; Lahfid et al. 2010; Aoya et al. 2010; Kouketsu et al. 2014; Lünsdorf et al. 2017). Raman-based metamorphic temperatures represent maximum metamorphic temperatures (Beyssac et al. 2002) as organic matter does not re-equilibrate during retrogression at lower temperatures in contrast to mineral assemblages used for conventional geothermometry.

The Kaczawa Metamorphic Complex (KMC) is a tectonostratigraphic unit composed mainly of Paleozoic low-grade metasedimentary and metavolcanic rocks, forming a tectonically complex structure with several folded thrust slices and mélange units. Metamorphism in the KMC ranges from low grade in mélange units to blueschist and later greenschist metamorphism (Baranowski 1975; Kryza et al. 1990, 2011; Baranowski et al. 1998; Kryza and Muszyński 2003; Kostylew 2008). The study presented by Kryza et al. (1990) indicated expected upper temperature limit for the regional metamorphism between 350 and 450 °C. The reported metamorphic temperatures obtained using various geothermometers on rocks from different units of the KMC vary from 150–200 °C through

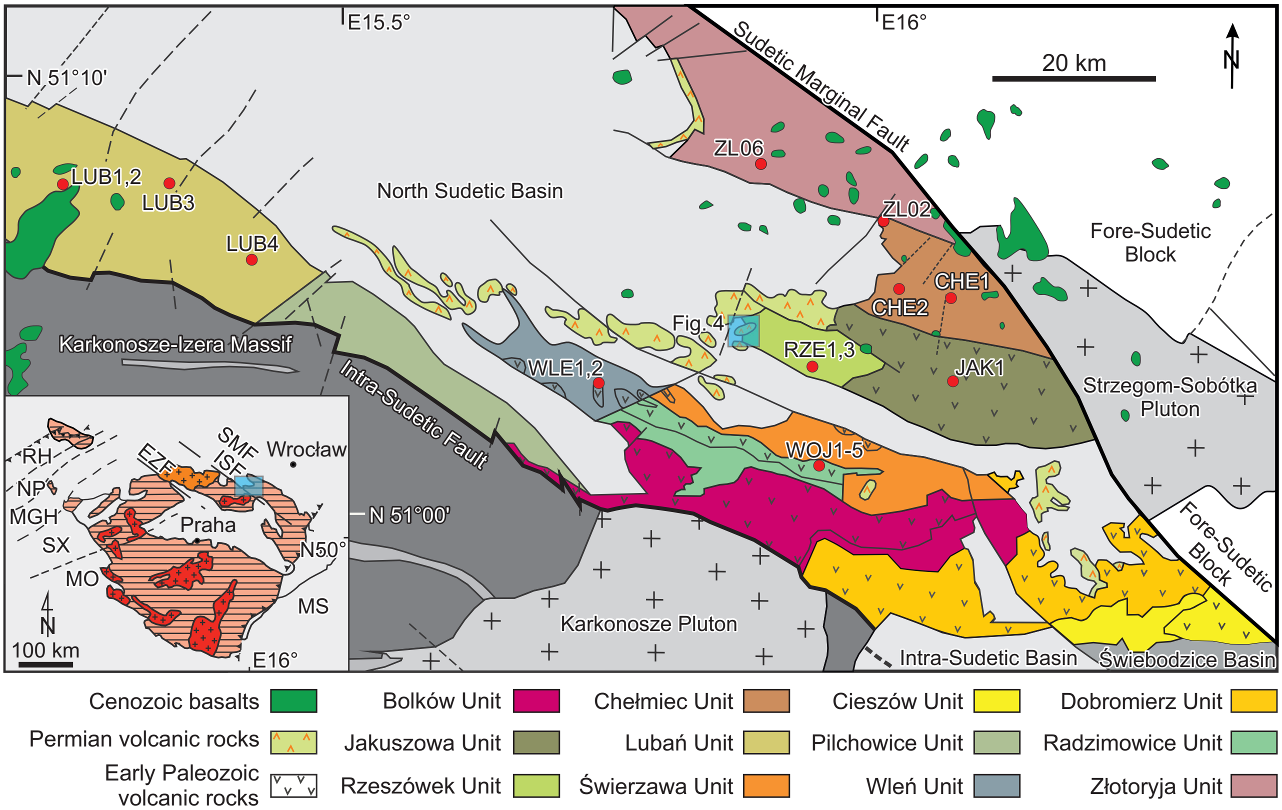

Fig. 1 Geological sketch map of the Kaczawa Metamorphic Complex and adjacent tectonic units (modified after Kryza and Muszyński, 1992; Cymerman, 2014). Sampling points are marked with red dots except for ROZ samples, which are marked on Figure 4. Inset maps show the location of the Kaczawa complex within the Bohemian Massif; EFZ – Elbe Fault Zone, ISF – Intra-Sudetic Fault Zone, MGH – Mid-German Crystalline High, MO – Moldanubian Zone, MS – Moravo–Silesian Zone, NP – Northern Phyllite Zone, RH – Rheno–Hercynian Zone, SX – Saxo–Thuringian Zone, SMF – Sudetic Marginal Fault, TB – Tepla–Barrandian Zone.

200–300 °C (Kostylew 2008), 270–310 °C (Kryza et al. 2011) to 350–470 °C (Baranowski 1975). This strongly suggests that the KMC preserves heterogeneous record of metamorphic temperatures in spatially close rocks. However, deciphering this record by using classical petrographic geothermometers would be problematic due to the lack of a single method that might be easily applicable to lithologically variable tectonic units. In addition, such comprehensive study would be also hampered by the thermal overprint from younger volcanic activity that locally modified or altogether reset the original metamorphic record. In order to identify such local anomalies, a large number of sampling sites would be required. It seems, however, that Raman-based geothermometry, being a relatively fast, robust and economic tool, could be used successfully for this purpose, especially that metasedimentary rocks with abundant carbonaceous material belong to the most widespread lithologies in the KMC.

The main aim of the study was to provide new and detailed information about the thermal history of the individual tectonic units within the KMC utilizing Raman-based geothermometry. In order to achieve this, we utilized several different Raman methods to obtain peak metamorphic temperatures. The precision and reliability of these methods is discussed, compared with previously published data, and their suitability is assessed as well. Additionally, the extent of influence of the contact metamorphism onto surrounding metasedimentary rocks, overprinting regional peak temperatures, was identified and evaluated.

Given the complicated tectono–metamorphic evolution, estimates of metamorphic temperatures in the KMC are surprisingly scarce and limited. Therefore, the addition of new geothermometric data and their extension to the units without previous temperature determinations will shed more light onto the development and tectonic evolution of the KMC in general and its individual units in particular.

2. Geological setting of the Kaczawa Metamorphic Complex

The study area is located within the Sudetes, NE part of the Bohemian Massif. The KMC is located in the West Sudetes, bordered by the possibly genetically related Görlitz Slate Belt (Aleksandrowski and Mazur 2002) to the W, the Izera–Karkonosze Massif to the SW, the Instrasudetic Basin and the Świebodzice Basin to the S, the Strzegom-Sobótka Massif to the S and SE, the Middle Odra Complex to the E and NE, and the North Sudetic Basin to the NW (Mazur et al. 2010). The Sudetic Boundary Fault (Aleksandrowski et al. 1997), which extends NW–SE, divides the KMC into a topographically distinct mountainous part in the SW and peneplained lowlands in the NE. The NE part of the KMC is mostly buried under younger Cenozoic strata, whereas the mountainous SW part (uplifted during the Neogene) is exposed at the surface as northern and southern branches separated by the North Sudetic Basin (Fig. 1). The North Sudetic Basin is a younger unit (Pennsylvanian to Late Cretaceous) that developed as an intramontane trough and evolved into a large depression filled with volcanic–sedimentary rocks, with the KMC lithologies forming its basement. The borders between the KMC and surrounding units are mostly developed as fault zones (Cymerman 2002). The fragmentary exposure and complex tectono–structural architecture of the highly folded and faulted rock sequence lead to difficulties in the interpretation of the geological evolution of the KMC. The KMC consists of Cambrian to Mississippian (Baranowski et al. 1990; Białek et al. 2007; Tyszka et al. 2008; Kryza et al. 2008; Jastrzębski et al. 2022, and references therein) metamorphosed sedimentary and volcanic succession with some evidence suggesting that the oldest KMC strata might date back to the late Precambrian (Kryza and Zalasiewicz 2008). The Paleozoic part of the KMC succession is represented by initial rift bimodal (felsic and mafic) volcanism together with metamorphosed shallow-water sedimentary rocks, followed by pelagic deposits and mid-ocean ridge basalts representing ocean deepening (Furnes et al. 1994; Kryza et al. 2007). The Variscan orogeny resulted in the formation of an accretionary prism and the development of mélange units that form the youngest part of the KMC (Baranowski et al. 1990, 1998; Collins et al. 2000; Kryza et al. 2007; Kryza and Zalasiewicz 2008).

Metamorphism and deformation during the Variscan orogeny resulted in intense faulting and thrusting, leading to the formation of several tectonic units with distinct burial and tectonic histories. The number of recognized separate tectonic units depends on the approach of the authors and varies from a few (Seston et al. 2000) to more than fifteen (Cymerman 2002). The tectonic model described by Seston et al. (2000) presents the Kowary shear zone at the base of the KMC as a decollement zone, with the Świerzawa thrust sheet (including the Świerzawa, Bolków, Złotoryja and Chełmiec units) resting on it. Overlying the Świerzawa thrust sheet is the Dobromierz thrust sheet (including the Dobromierz, Rzeszówek, and Jakuszowa units), with the Kaczorów shear zone separating the two thrust sheets. According to the model of Cymerman (2002), the furthest NW Lubań and Bolesławiec units, represent a para-autochthon of the continental margin. The para-autochthon is covered by metasedimentary–metavolcanic allochthonous units (Świerzawa, Wleń, Bolków, Radzimowice, Pilchowice, Rzeszówek, Chełmiec, Złotoryja), which were thrust onto the para-autochthon to the west. The allochthonous units were also overthrust by the predominantly MORB-type metavolcanic Dobromierz and Jakuszowa units.

During the late Paleozoic development of accretionary prism, the KMC succession was subjected to high-pressure blueschist metamorphism followed by the greenschist facies stage (Baranowski 1975; Smulikowski 1990, 1995; Kryza et al. 1990, 2011). The most detailed metamorphic paths were presented by Kryza et al. (2011) for the southern branch of the KMC. Based on the mineral assemblages and assuming chemical equilibrium between mineral phases, they recognized four metamorphic stages: high pressure stage I at 8.5 ± 0.5 kbar and 270 ± 203 °C in a subduction-related environment, thermal relaxation in stage II at 6.0 ± 1.0 kbar and 310 ± 20 °C, cooling and decompression at stage III at 3.5 ± 0.5 kbar and 280 ± 20 °C, followed by stage IV at <3.5 kbar and <280 °C. Also, several other attempts have been made to determine metamorphic temperatures in the KMC rock sequence. Kryza et al. (1990) established an upper limit for the greenschist facies metamorphism between 350 and 450 °C based on the mineral assemblage of metabasites from the Świerzawa unit. Kostylew (2008) estimated the metamorphic grade of mélange, turbidite and metamudstones based on the illite crystallinity index as anchizone for the Rzeszówek unit and some rock types of the Chełmiec unit with a temperature range between 200 and 300 °C and late diagenesis for a part of the turbidite sequence from the Chełmiec unit with a temperature range of 150–200 °C.

The blueschist high-pressure metamorphism has been only recognized in the southern branch of the KMC (Świerzawa unit, Kryza et al. 1990, 2011) and to the present authors knowledge there is no indication of it in the northern branch. The lack of high-pressure metamorphism record in the northern branch may be the result of an overprint of later metamorphic conditions, but it could also suggest that the northern and southern KMC branches experienced different metamorphic conditions.

Apart from regional metamorphism, some areas of the KMC were subjected to local thermal metamorphism caused by volcanic and sub-volcanic bodies of various sizes. Late- and post-orogenic Variscan volcanism (Awdankiewicz 2006; Mazur et al. 2007; Awdankiewicz et al. 2014, and the references therein) resulted in the formation of extensive bodies in the overlying North Sudetic Basin with smaller bodies located within or at the contact with the KMC lithologies. Another volcanic event in the area occurred in the Paleogene and Neogene (Pańczyk et al. 2023, and the references therein) as part of the activity in the Central European Volcanic Province, which was associated with continental rifting (Dèzes et al. 2004; Ulrych et al. 2011). Particularly rich in volcanic bodies are the western (Lubań unit) and northeastern (Złotoryja and Chełmiec units) parts of the KMC, including the lowland NE segment of the KMC.

3. Samples and analytical conditions

3.1. Samples and sampling strategy

Twenty-five samples of dark grey to black Ordovician and Silurian metasedimentary rocks were collected from seven different units of the KMC: Lubań, Wleń, Radzimowice, Rzeszówek, Jakuszowa, Chełmiec and Złotoryja (Fig. 1). Sampling sites were selected based on lithology information from the geologic maps and available outcrops descriptions. Sampled lithologies were described in the literature as quartzites, graphitic slates, graptolitic slates, black slates, grey slates, siliceous slates, clay phyllites or (meta)lydites (Baranowski 1975; Kryza and Muszyński 1992; Collins et al. 2000; Kryza et al. 2008, 2011). They were interpreted as metamorphosed fine-grained sedimentary rocks, originally oceanic pelagic (Baranowski 1975; Baranowski et al. 1990) or hemipelagic (Baranowski et al. 1998) deposits. All the samples collected contain organic matter that occurs in three forms: dispersed fine particles, fossilized graptolites or other zooclasts, and secondary solid bitumen. The dominant form is fine-grained dispersed organic matter with roughly micrometer size individual grains either uniformly dispersed or concentrating within organic-rich laminae (Fig. 2a, b). Several samples contain fossilized graptolites, visible either with the naked eye on the cleavage surfaces or in thin-sections (Fig. 2a, c). When the foliation is recognizable in the thin sections, the graptolites are aligned parallel to foliation, reaching several hundred micrometers in size. In some of the samples secondary solid bitumen was also present filling voids in recrystallized matrix of the rock (Fig. 2b, d).

To estimate a possible impact of thermal overprint caused by volcanic activity postdating regional metamorphism in the KMC a special attention was paid to sampling near the exposure of the Wielisławka Rhyolite in the Rzeszówek unit. The Wielisławka Rhyolite is a 0.4×1.2 km body, representing a gently inclined plug or an inner part of a lava dome (Awdankiewicz and Szczepara 2009). The rhyolite was dated at 292.8 ± 2.1 and 297.5 ± 2.8 Ma (Mikulski and Williams 2014; Awdankiewicz et al. 2014) being younger than the surrounding lower Paleozoic sampled formations. The occurrence of organic matter-rich metasedimentary rocks at varying distance from the Wielisławka Rhyolite, starting from the contact zone itself up to a distance of about 500 m away, allows to recognize samples affected or not affected by contact metamorphism. It was assumed that once the influence of thermal metamorphism on the measured spectral parameters was established, it could be used to detect the same local overprint in regions where the relationship between sampling points and volcanic bodies is not clear.

3.2. Raman measurement and spectra processing

Samples were prepared as standard petrographic thin sections, oriented perpendicular to foliation, if it was recognizable. Raman spectra were acquired at the Institute of Geological Sciences, University of Wrocław, Poland using a Renishaw inVia Qontor spectrometer integrated with a Leica DM2700 confocal microscope equipped with a Renishaw Centrus CCD detector. The 532 nm frequency laser with a spot size approximately of 1 µm and 0.09 mW laser power at the sample surface was used with a 50x objective. The measurements were performed on organic matter located below the thin section's surface to avoid polishing effects (Wopenka and Pasteris 1993; Lünsdorf 2016; Kouketsu et al. 2019). Acquisition times varied between 50 and 200 seconds depending on the signal intensity. Internal calibration, using a standard Si wafer, was performed prior to spectra acquisition. Ten spectra per sample in the range 900–2000 cm^{-1} were collected, processed and used for calculation of metamorphic temperatures. The spectra were processed using the Renishaw's WiRE 5.3 software.

Metamorphic temperatures were calculated according to the four different methods further referred to as Rahl (Rahl et al. 2005), D1 and D2 (Kouketsu et al. 2014), and IFORS (Lünsdorf et al. 2017) methods. The methods process the spectra in different ways, including spectra deconvolution into a different number of component curves. Figure 3 presents example of spectrum (sample ZLO 6) deconvolution according to the three methods applied in this study.

For the Rahl method, the spectra were subjected to baseline subtraction. As Rahl et al. (2005) did not provide guidelines for the baseline subtraction, the spectra were subjected to a 3-rd order polynomial baseline subtraction as suggested by Henry et al. (2018) for the samples with fluorescence. Subsequently, the spectra were deconvoluted (Fig. 3a) into D1 (1350 cm^{-1}), D2 (1620 cm^{-1}), D3 (1510 cm^{-1}), and G bands (1580 cm^{-1}) and R1 and R2 parameters (supplementary material) were calculated as:

$R1 = (D1/\mathrm{G})_H$, where H is the height of the bands

$R2 = [D1/(\mathrm{G} + D1 + D2)]_A$, where A is the area of the bands. Metamorphic temperatures were obtained according to the following equation:

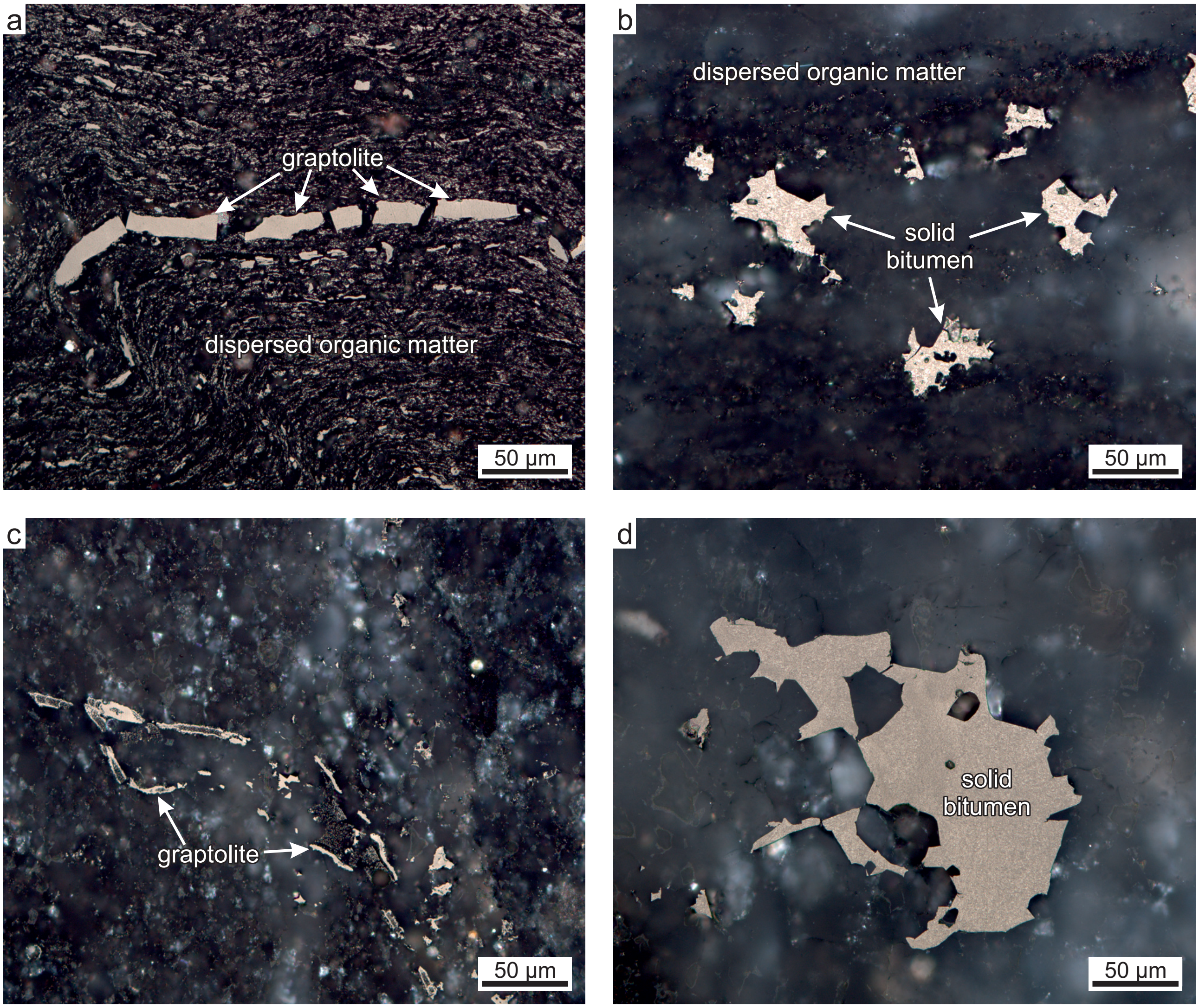

Fig. 2 Photomicrographs representing different types of organic matter visible in studied samples (reflected light, oil immersion): **a** – organic matter is evenly dispersed through the sample, visible as light grey particles aligned parallel to the foliation; larger particles in the center represent graptolite remnant; sample ROZ2, **b** – small dark particles of dispersed organic matter concentrate in bands parallel to foliation; larger particles represent migratory solid bitumen filling voids between mineral grains, sample WOJ5, **c** – disjointed graptolite remnant is visible in the center; small, dark particles of organic matter are visible dispersed through the picture; sample ZLO4, **d** – large migratory solid bitumen visible in the center filling voids between mineral grains; euhedral crystal shapes are outlined by surrounding solid bitumen; sample ZLO4.

$$T(°C) = 737.3 + 320.9\,R1 - 1067\,R2 - 80.638\,R1^2$$

For the Kouketsu method, the spectra were subjected to linear baseline correction in the 1000–1750 cm^{-1} range followed by deconvolution (Fig. 3b) into D1, D2, D3, D4 and G bands (Fitting E and F). Full widths at half maximum (FWHM) from D1 and D2 bands (supplementary material) were obtained and metamorphic temperatures were calculated according to the following equations:

$$T(°C) = -2.15(FWHM-D1) + 478$$

$$T(°C) = -6.78(FWHM-D2) + 535$$

The IFORS software was proposed by Lünsdorf and Lünsdorf (2016) and Lünsdorf et al. (2017) to improve the comparability of Raman results avoiding bias resulting from users' interpretation during the spectra fitting process. The software curve-fits Raman spectra (Fig. 3c) modelling at the same time the spectrum fitting by using pseudo-Voight bands together with the background subtraction (performed with a fifth-order polynomial curve). The number of curves for deconvolution is not fixed like in the two previous methods. This iterative process runs until the best representation of the baseline-subtracted spectrum is reached (Lünsdorf and Lünsdorf 2016; Lünsdorf et al. 2017). The Raman parameters derived from this process are listed in the supplementary materials and include the

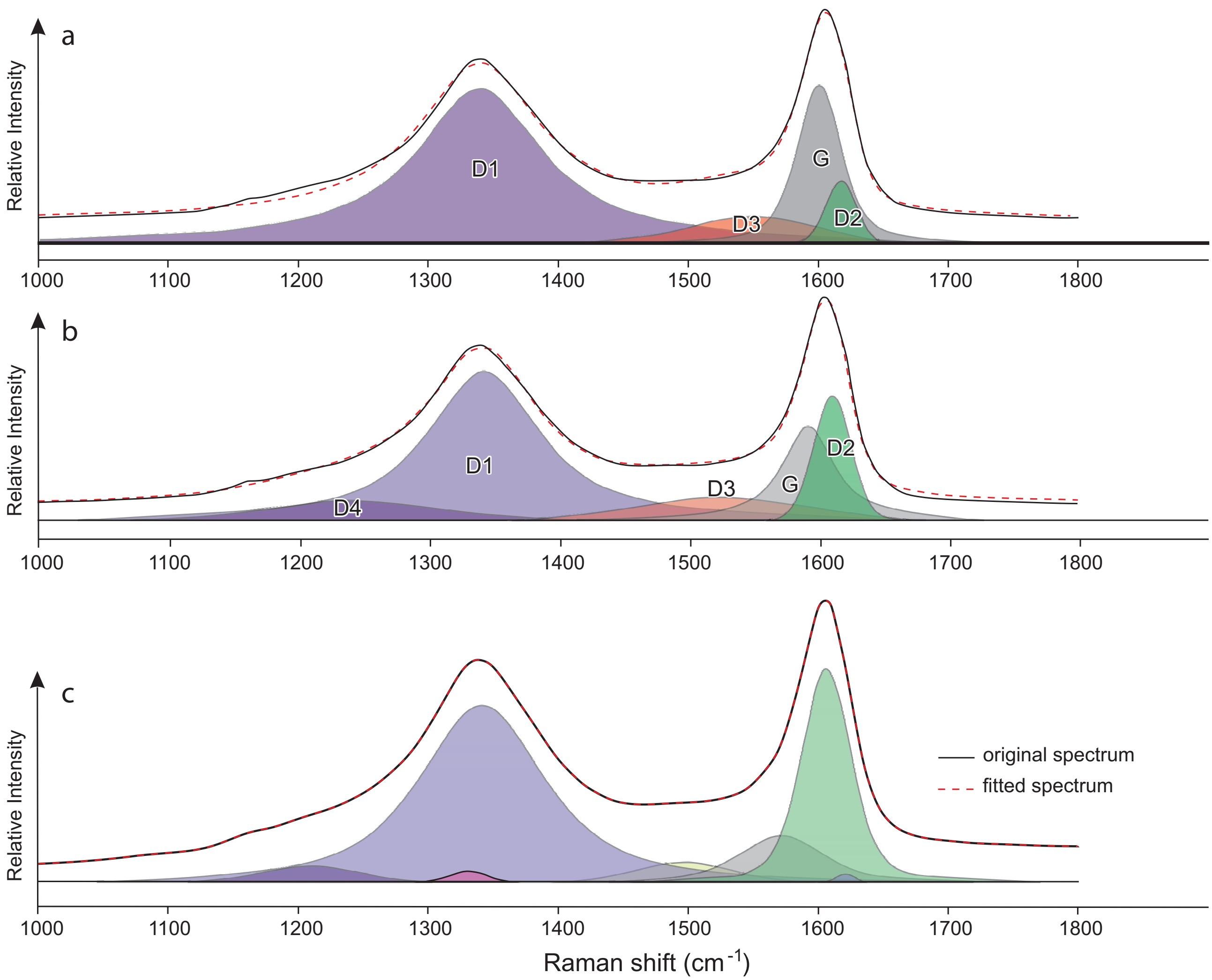

Fig. 3 Example of spectrum deconvolution (sample ZLO6) according to: **a** – Rahl, **b** – Kouketsu, and **c** – IFORS methods.

normalized intensities of the D and G bands (STA-D, STA-G), the G-shape-factor, which depends on the presence and intensity of the D2 bands (Lünsdorf and Lünsdorf 2016), the mean position of the D and G bands and their intensity ratio. The STA-D, STA-G are used to calculate the temperature. The calibration curve provided in Lünsdorf et al. (2017) is based on a series of 26 samples with the record of metamorphic temperatures between 100 and 700 °C.

Errors associated with calibration of Raman methods to conventional mineralogical geothermometers are ± 30 °C for D1, 50 °C for D2, 30 °C for IFORS, and 50 °C for Rahl as presented in papers detailing the methods (Rahl et al. 2005; Kouketsu et al. 2014; Lünsdorf et al. 2017). These should be considered as absolute temperature errors. The actual standard deviations presented in this paper (Tab. 1) are much smaller and indicate that relative temperature errors between the samples are much smaller.

Apart from the metamorphic temperatures, Raman band separation (RBS) distances were also measured. The RBS is a parameter decreasing its value with increasing metamorphic temperatures in the studied temperature range (Henry et al. 2019).

4. Results

Calculated metamorphic temperatures and RBS (averages of 10 spectra per sample) are presented in Tab. 1 (for individual temperatures and RBS values obtained from each collected spectrum please see supplementary material). Overall calculated temperatures range between 200 and 377 °C, with differences between different tectonic units and between different calculation methods. Two of the methods used (D1 and IFORS) provide peak temperatures that are similar to each other. The Rahl and D2 methods provided on average lower temperatures than D1 and IFORS, with Rahl providing the lowest temperatures of the applied methods. Some of the highest obtained temperatures are potentially from samples overprinted by local contact metamorphism, and these samples will

Tab. 1 Average metamorphic temperatures according to the different methods and Raman band separation (RBS) obtained for the samples from the Kaczawa Metamorphic Complex. Samples possibly affected by contact metamorphism in *italics*.

Tectonic Unit	Sample name	Age	Rahl T [°C]	±	D1 T [°C]	±	D2 T [°C]	±	IFORS T [°C]	±	RBS [cm^{-1}]
Złotoryja	ZLO6	Silurian	200	7	248	24	313	8	278	6	261.6
	ZLO2	*Silurian*	*361*	*8*	*353*	*6*	*278*	*26*	*357*	*7*	*250.1*
Chełmiec	*CHE1*	*Silurian*	*293*	*28*	*322*	*10*	*223*	*10*	*325*	*9*	*254.5*
	CHE2	Silurian	236	10	299	9	296	19	301	9	261.2
Jakuszowa	JAK1	Silurian	201	7	278	5	313	6	283	4	265.7
Rzeszówek	RZE1	Ordovician	214	8	291	6	308	7	291	4	263.8
	RZE3		211	9	292	7	305	5	289	5	263.7
	ROZ1	Silurian possibly Ordovician	225	34	280	9	323	9	289	4	261.5
	ROZ2		225	29	303	11	313	6	296	9	263.5
	ROZ3		*265*	*28*	*252*	*11*	*338*	*5*	*274*	*9*	*255.4*
	ROZ7		254	36	271	5	320	6	286	9	261.6
	ROZ8		246	36	276	12	323	5	288	9	262.6
	ROZ13		*296*	*23*	*365*	*3*	*299*	*47*	*349*	*16*	*251.2*
	ROZ15		*268*	*9*	*295*	*17*	*330*	*43*	*290*	*24*	*258.0*
Wleń	WLE1	Silurian	311	24	353	13	308	50	347	17	251.4
	WLE2		270	21	310	18	267	16	314	17	254.7
Radzimowice	WOJ1	Silurian	271	6	320	7	212	11	322	12	253.1
	WOJ2		267	9	322	10	220	16	320	14	254.2
	WOJ3		275	8	343	15	211	16	346	18	252.1
	WOJ4		276	8	344	10	215	20	345	12	252.8
	WOJ5		270	7	331	16	235	31	326	15	254.3
Lubań	*LUB1*	*Silurian*	*321*	*19*	*352*	*11*	*273*	*31*	*348*	*11*	*248.9*
	LUB2		*347*	*8*	*377*	*2*	*292*	*10*	*375*	*11*	*245.2*
	LUB3	Silurian	254	11	306	5	270	13	312	8	257.4
	LUB4	Ordovician	348	8	345	4	276	5	352	6	251.5

be discussed separately in section 5.1. Peak temperatures for the regional metamorphism (excluding contact metamorphism samples) according to IFORS method range between 278 and 352 °C. The units located in the southern branch of KMC (Wleń, Radzimowice, and Lubań) are characterized by higher temperatures (314–352 °C range) than units (Złotoryja, Chełmiec, Jakuszowa, Rzeszówek) from the northern branch (278–301 °C). Similar results are obtained from D1 method with temperature ranges 306–353 °C for the southern and 248–303 °C for the northern units. The results from the D2 method generally show more scatter with temperatures 211 to 308 °C for the southern units and 296 to 323 °C for the northern units. The lowest peak temperatures are obtained using Rahl method with 254–348 °C range for the southern units and 200–254 °C range for the northern units. The differences between applied methods will be further discussed in section 5.2.

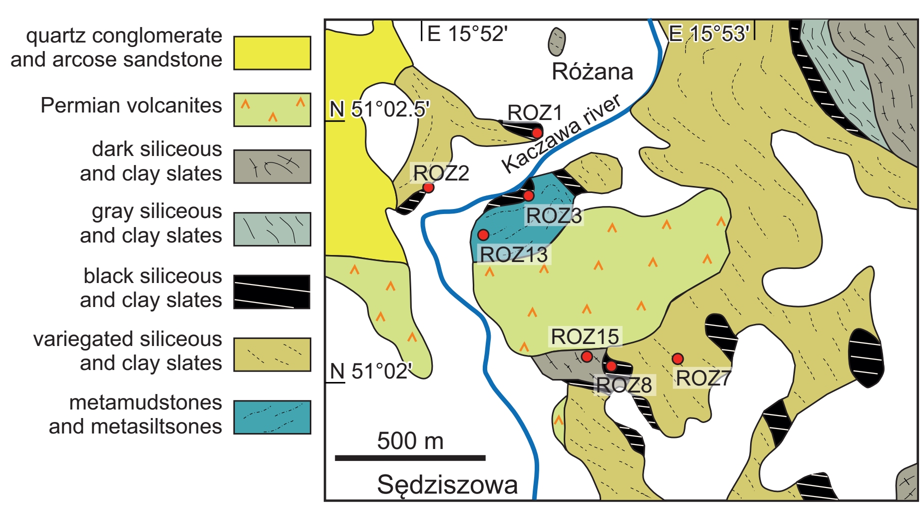

Fig. 4 Location of the sampling points near the Wielisławka Rhyolite (modified after Haydukiewicz 1977).

5. Discussion

5.1. Influence of contact metamorphism

The influence of contact metamorphism was tested in the area where the studied metasedimentary rocks are exposed in the vicinity of a younger volcanic body of the Wielisławka Rhyolite in the Rzeszówek unit (Fig. 4). Seven samples were collected within 500 m distance from the rhyolite (Fig. 4, Tab. 1, samples with prefix ROZ) and yielded spectroscopic signals from organic matter that was strong enough to calculate metamorphic temperatures. To estimate the influence of contact metamorphism, the RBS measured in the spectra was compared with the distance from a sampling point to the contact with the Wielisławka Rhyolite (Fig. 5). The RBS parameter was chosen because it is the least affected by the spectral processing procedure of all calculated spectral parameters. The distance to the rhyolite was measured from geological maps and may differ slightly from the true distance. However, this should not significantly affect the results. The samples collected 150 m or further from the rhyolite, with RBS about 260–265 cm^{-1}, do not show changes in RBS with increasing distance from the volcanic body. This suggests they are not affected by contact metamorphism and their Raman parameters are representative of regional metamorphism. The two samples on the Fig. 5 with the furthest distance from the volcanic body (RZE1 and RZE3) are also from the same Rzeszówek unit with about 5 km distance from the rhyolite. They were taken closer (350 and 300 m, respectively) to a smaller ca. 100 × 150 m Neogene volcanic body (Frąckiewicz 1958) and based on the distance to the closest volcanic body should not be affected by contact metamorphism. They can serve as reference points, with the thermal maturity of the organic matter assumed to result solely from regional metamorphism. This is corroborated by the RBS values between 260 and 265 cm^{-1} similar to the ROZ samples collected with 150 m or more distance from the Wielisławka Rhyolite. The three samples taken within 100 m distance of the rhyolite (ROZ3, ROZ13, and ROZ15) show significant decrease in the RBS (Fig. 5). Thus, it can be concluded that the contact metamorphism aureole around the Wielisławka Rhyolite extends up to 100 m away from the rhyolite. Similar values for subvolcanic bodies of comparable size were reported in the literature [e.g. 110 m by Mori et al. (2017)]. In general, it seems that a significant decrease in the RBS values in a population of samples from a geological unit with a homogeneous record of regional metamorphism conditions may effectively reflect a local thermal overprint.

Apart from the samples from the vicinity of the Wielisławka Rhyolite, some other samples were also collected within a few hundred m distance from volcanic bodies (Tab. 1). The sampling point for CHE1 sample is located within 50 m distance from tufoids and 150 m from a diabase body, both of unknown age (Jerzmański 1954), whereas CHE2 was sampled far away from any volcanic bodies. Both samples represent the Chełmiec unit. Sample CHE1 is characterized by smaller RBS value and higher calculated metamorphic temperatures than CHE2, possibly indicating that the volcanic bodies are younger than the sampled Silurian quartzite and thermally affected CHE1. Sample of Silurian silica schist ZLO2 (Złotoryja unit) was located within 150 m of the metavolcanites considered Ordovician to Devonian in age (Jerzmański and Walczak-Augustyniak 1995). Again, the RBS value is lower than in the other sample from the same unit (ZLO6) that was obtained from an outcrop with no volcanic bodies nearby. This also suggests that the ZLO2 sample is possibly affected by contact metamorphism and the metamorphosed volcanic rocks are younger than the Silurian silica schists. The samples LUB1 and LUB2 from the Lubań unit were collected at a 200 m distance from current exposure of a Neogene basaltic body (Berezowski 1959). As the samples show slightly smaller RBS values than other samples from the Lubań unit (LUB3 and LUB4) they are possibly located within the contact aureole. All samples with such anomalously low RBS values and similarly reduced calculated metamorphic temperatures are marked in Tab. 1 as possibly affected by local contact overprint. Such estimates can hardly be taken as evidence for thermal metamorphism, and each of these cases should be verified by more detailed studies and analyses, similar to those performed for the area near the Wielisławka Rhyolite. However, they have not been included in the discussion of regional metamorphic temperatures.

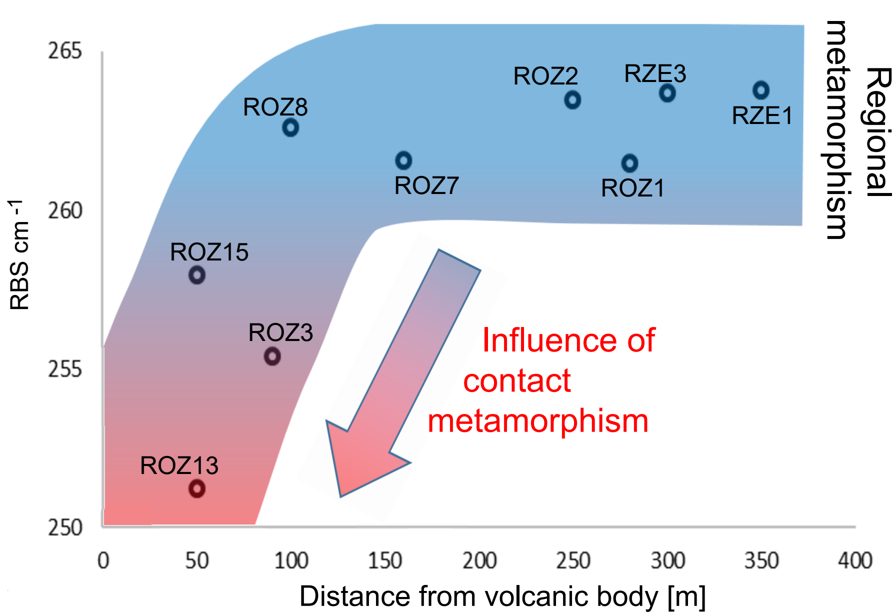

Fig. 5 Influence of contact metamorphism on the Raman band separation (RBS) with the distance from volcanic body.

Tab. 2 Temperature differences (in absolute numbers) between the four different methods applied for the samples from different tectonic units of the Kaczawa Metamorphic Complex. Samples possibly affected by contact metamorphism in *italics*.

Tectonic Unit	Sample name	Temperature Difference [°C]					
		Rahl-D1	Rahl-D2	Rahl – IFORS	D1-D2	D1-IFORS	D2-IFORS
Złotoryja	ZLO6	48	112	77	65	30	35
	ZLO2	*9*	*83*	*4*	*74*	5	79
Chełmiec	*CHE1*	*29*	*71*	*31*	*99*	3	102
	CHE2	64	61	66	3	2	5
Jakuszowa	JAK1	77	112	82	35	5	30
Rzeszówek	RZE1	78	95	77	17	1	18
	RZE3	81	93	78	12	3	16
	ROZ1	54	97	63	43	9	34
	ROZ2	79	88	72	10	7	16
	ROZ3	*12*	*73*	*9*	*86*	21	64
	ROZ7	17	66	32	49	15	34
	ROZ8	30	77	41	47	12	35
	ROZ13	*68*	*3*	*53*	*66*	15	51
	ROZ15	*27*	*61*	*22*	*35*	5	40
Wleń	WLE1	42	3	36	45	7	38
	WLE2	40	4	43	44	3	47
Radzimowice	WOJ1	49	59	51	108	2	110
	WOJ2	54	47	53	102	1	100
	WOJ3	67	64	70	131	3	134
	WOJ4	67	61	68	128	1	129
	WOJ5	62	35	57	97	5	92
Lubań	*LUB1*	*30*	*48*	*27*	*79*	3	75
	LUB2	*30*	*55*	*28*	*85*	2	83
	LUB3	51	16	57	36	6	41
	LUB4	3	72	4	69	7	76
average		47	62	48	63	7	59
maximum		81	112	82	131	30	134

5.2. Comparison between different Raman methods and previously reported temperatures

In the discussion concerning metamorphic temperatures achieved during regional metamorphism only samples with no apparent overprint due to contact phenomena were considered (Tab. 1). Metamorphic temperatures have been calculated in this paper according to four different methods (Rahl et al. 2005; Kouketsu et al. 2014; Lünsdorf et al. 2017) showing some differences. The average differences between the pairs Rahl-D1, Rahl-D2, D1-D2, D2-IFORS, Rahl-IFORS are presented in Tab. 2 as absolute numbers. The differences vary by over 50 °C, with several individual differences exceeding 100 °C, which indicates significant discrepancies between the methods. There are two methods, however which give very similar results. The pair D1-IFORS shows an average of the differences equal to 7 °C, with the largest individual difference of 30 °C, and 80 % of the differences are less than 10 °C. Given the fact that the two methods are based on different Raman parameters, D1 being based on the band width and IFORS based on the area of selected peaks, their high correlation suggests that the methods are well calibrated for the investigated temperature range.

Both D2 and Rahl methods give on average lower temperatures than D1 and IFORS. A similar discrepancy was observed by Lünsdorf et al. (2017) with Rahl's being lower than IFORS temperatures, especially below 400 °C. Croce et al. (2020) also compared D1 and Rahl methods, concluding that the latter is less reliable due to the higher variability of the values obtained. As for the D2 parameter, Kouketsu et al. (2014) noted that the error associated with D2 is ± 50 °C, while the error associated with D1 is around ± 30 °C indicating that D1 is more reliable than D2.

Data on metamorphic temperatures of the KMC rocks are very scarce in general and no such data exists for rocks from outcrops close enough to the sampling points of this study to be used as direct reference data. To the best of the present authors' knowledge, the geographically closest are the maximum metamorphic temperatures of 310 °C presented by Kryza et al. (2011), which were determined for the metavolcanic rocks 2 km from WOJ samples of the Wojcieszów unit. The WOJ samples yielded Raman-based temperatures that clustered closely together, giving on average 219 °C based on D2, 272 °C

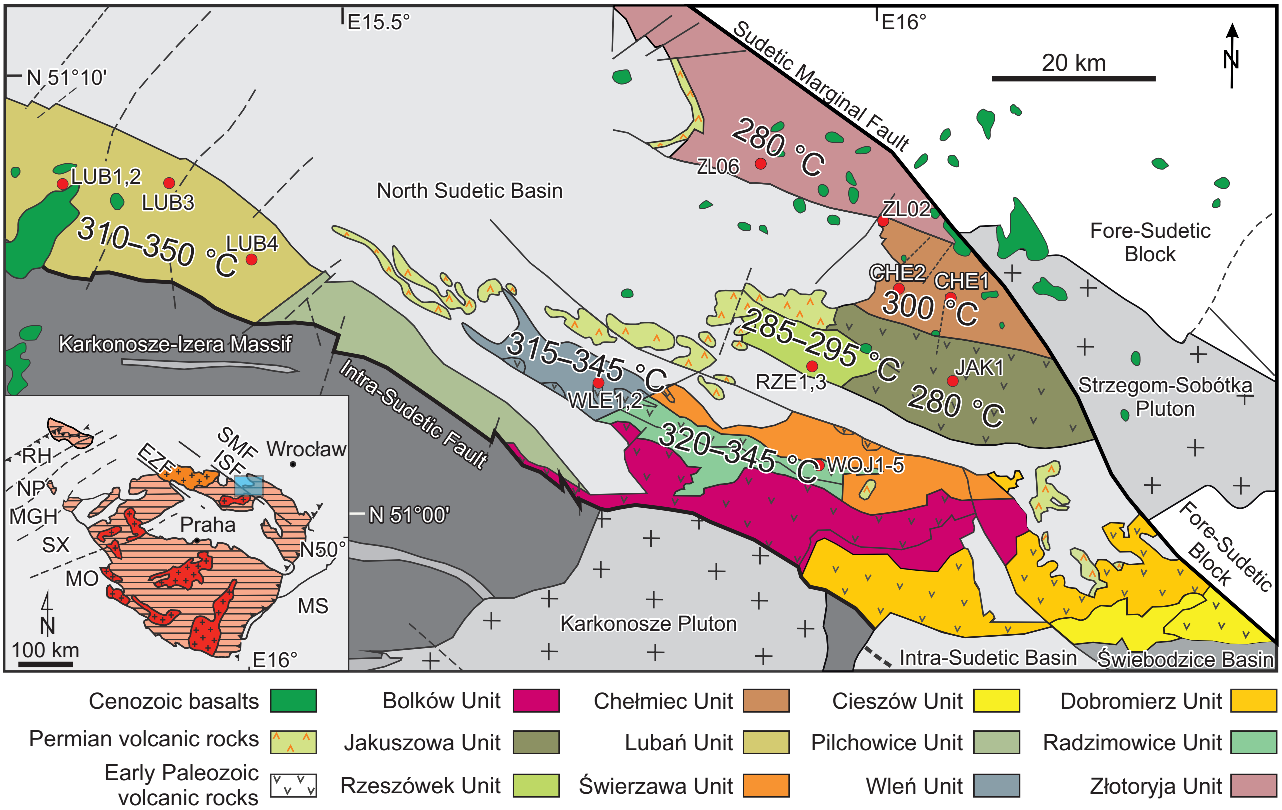

Fig. 6 Geological sketch map of the Kaczawa Metamorphic Complex and adjacent tectonic units (modified after Kryza and Muszyński, 1992; Cymerman, 2014) with obtained regional metamorphism maximum temperature ranges.

based on Rahl, and 332 °C based on both D1 and IFORS, with the latter two closest to previously reported temperatures (Kryza et al. 2011). In can be concluded that the IFORS and D1 methods are the most applicable for calculating temperatures of regional metamorphism in the studied temperature range.

5.3. Insights into the KMC regional metamorphism

The temperatures obtained by the D1 and IFORS methods are very close to each other. However, the IFORS data are used in discussing regional implications because this method is not affected by user bias (Lünsdorf et al. 2017) and provides the closest agreement between fitted and original spectra (example in Fig. 3). Nonetheless, the discussion would be almost the same if based on the D1 temperatures, and even if based on Rahl's temperatures, would show similar variance between different units of the KMC, although with lower peak metamorphic temperatures.

The obtained peak metamorphic temperatures for the KMC (278 to 352 °C, IFORS method; Tab. 1, Fig. 6) agree well with the temperatures previously obtained by Kryza et al. (2011), confirming that the KMC experienced temperatures in the lower range of blueschist and greenschist metamorphism. The tectonic models presented by Seston et al. (2000) and Cymerman (2002, 2014) suggest that the KMC is composed of several thrust sheets. The thrust direction was toward NW, and the SE units are thrust onto the NW units. The temperatures obtained in this study for several different units show no gradient from the NW to SE, indicating similar metamorphic conditions for the different stacked thrust sheets.

When comparing the peak metamorphic temperatures across the seven KMC units (Fig. 6, Tab. 1) it is evident that the units located in the southern branch exhibit higher regional metamorphic temperatures than the units located in northern branch of the KMC. The temperatures calculated for the southern branch (the Lubań, Wleń, and Radzimowice units) range from 312 to 352 °C with the average temperature of 331 °C. The temperatures calculated for the northern KMC branch (the Chełmiec, Jakuszowa, Rzeszówek, and Złotoryja units) range from 278 to 301 °C with the average temperature of 289 °C. The southern and northern KMC branches are separated on the surface by the Permian–Mesozoic North Sudetic Basin; a synclinal structure extending along the SE–NW axis with fault boundaries between the basin and exposed parts of the KMC (Solecki 1994; Głuszyński and Aleksandrowski 2022 and references therein). Both the North Sudetic Basin and underlying KMC are cut by numerous faults with both vertical and horizontal displacement (Bałazińska and Bossowski 1979; Kowalski 2021; Głuszyński and Aleksandrowski 2022). However, there is no indication that any of the KMC branches was significantly uplifted relatively to the other side. The difference in peak metamorphic temperatures suggests that the metamorphic conditions for the southern branch were somewhat different with the peak temperatures on average 40 °C higher than for the north branch. Based on the conditions of 310 °C and 6 kbar, during the peak metamorphic temperatures obtained by Kryza et al. (2011) we can estimate geothermal gradient at that time to be close to 15 °C/km. Thus, the difference in peak temperatures between the two KMC branches suggests that the southern branch was buried 2.5–3.0 km deeper than the northern KMC branch. The fact that peak temperatures are different between the northern and southern branches rather than between the different thrust sheets as modeled by Seston et al. (2000) and Cymerman (2002, 2014) also indicates that peak metamorphic temperatures were reached after the thrust sheet stacking has already taken place.

6. Conclusions

In this study, the Raman geothermometry was utilized to constrain the thermal history of the KMC. Some of the obtained peak temperatures were suspected to be affected by thermal overprint from contact metamorphism. The RBS was used to assess the extent of the contact aureole for the Wielisławka Rhyolite; one of the largest volcanic bodies within the KMC. Based on the size of the contact aureole (estimated to be 100–150 m) some samples were excluded from the assessment of regional metamorphic peak temperatures. Four different geothermometry methods (Rahl et al. 2005; Kouketsu et al. 2014; Lünsdorf et al. 2017) were applied. Two of the methods (IFORS and D1) gave equivalent results, comparable with previously reported temperatures and are considered more reliable. The Rahl method indicated similar trends, but the calculated temperatures were lower than those of IFORS and D1.

The regional metamorphic temperatures for seven different tectonic units (278 to 352 °C range) correspond to the temperature range for the blueschist and greenschist metamorphic facies, as previously described (Baranowski 1975; Smulikowski 1990, 1995; Kryza et al. 1990, 2011). There is no metamorphic temperature gradient along the NW–SE extent of the KMC, indicating that the different thrust sheets within the KMC experienced similar metamorphic conditions. However, a temperature difference between the northern (278 to 301 °C) and the southern (312 to 352 °C) branches of the KMC indicates slightly different metamorphic conditions between the branches. The difference of 40 °C in peak metamorphic temperatures suggests that the southern branch was buried 2.5–3.0 km deeper than the northern branch.

Raman-based geothermometry was successfully used to decipher metamorphic temperatures, especially that carbonaceous metasedimentary rocks are among the most widespread lithologies in the KMC. It revealed previously unknown differences in peak metamorphic temperatures between the northern and southern KMC branches, providing insight into the evolution of the KMC, which should be considered when reconstructing the regional metamorphic evolution of the Sudetic segment of the European Variscides.

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