Dear Dr. Rahl,

Thank you very much for taking the time to review our manuscript and for your highly constructive feedback – your comments have been extremely useful in helping to refocus our paper. As outlined in our response to A. Jubb, we have made the following significant changes:

- We have focussed the paper on the changes to individual Raman spectral parameters rather than geothermometric equations, only including the geothermometry as a minor discussion point later on in the text. We believe that this makes the manuscript far clearer and more focussed, not to mention easier for the reader to surmise the key points of the study.
- Transects across faults and shear zones are presented in more detail.
- Error ranges have been added to both text and figures.
- Methodology and definitions have been significantly tightened.

As per your suggestion, we have introduced a new figure which includes detailed transects of each locally strained locality, showing the precise changes exhibited by each spectral parameter (Figure 6). We have ultimately focussed the results and discussion around this figure, which we believe has brought greater clarity and focus to the manuscript. Having included this extra detail, we made the decision to omit the section dedicated to various geothermometers, as it was evident that this was confusing to the reader and that this section may be worthy of a study in itself. A future paper which dealt with the geothermometry section alone would be capable of doing greater justice to this complex subject, the scope of which would include all the different methodologies associated with specific geothermometers. In the meantime, the revised manuscript focusses in greater detail on the fine-scale transects, as you suggest, with a discursive note on the Schito and Corrado equation as an implication of our findings rather than an induvial set of results. By centring our results around the individual transects, we have additionally been able to discuss potential strain gradients. These strain gradients, and our definitions of 'strained' vs. 'background', are now explained in greater detail in lines 99-124, where we have included additional information. Please note that due to the different lithologies involved in each transect, it was more difficult to quantify the relative strain than in (for example) Kedar et al (2020) where the entire shear zone was contained within one lithology.

Thank you also for your specific comments, which have been addressed individually below. We have added error bars to our figures and specified them appropriately in the text, and have investigated where there are inconsistencies such as those highlighted in your comment above. Additionally, we have emphasised our discussion on other factors which may influence the changes observed in Raman spectra, also as per your suggestions above. Once again, we are grateful for your feedback and believe that this has greatly helped us to improve the manuscript.

Yours sincerely,

L. Kedar, C. E. Bond, and D. Muirhead.

Specific comments

Line 89 – "background" strain? Given the purpose of this study, assessing and describing this "background" strain is important. Can the strain in these rocks be quantified?

We agree that this could be better defined, and have therefore added a further few paragraphs (Lines 91-111) to explain the categorisation in more detail. It was harder to quantify the strain levels in the rocks than, for example, in our 2020 paper (Carbon ordering in an aseismic shear zone: implications for Raman geothermometry and strain tracking, Kedar et al., 2020), where the shear zone in question occupied a single lithological unit. In the current manuscript, several faults and shear zones involve multiple lithologies, and therefore most sampling was based on judgement in the field. This strategy is better explained in the additional text mentioned above.

Line 95 – one sample or a cluster? I don't understand what was done in practice here. How closely spaced were samples in a "cluster"? How were they averaged together? (Physically, or were analytical results averaged?)

If multiple samples were collected at an outcrop and these showed similar analytical results, these results were averaged.

Line 190 – I am confused on the analytical procedure used here. Above (line 185) it says that for each sample, 10 grains were studied, and that each grain was scanned three times. That would make 30 analyses per sample. Yet here it says "this process was carried out 3 times for each acquired spectrum, resulting in 30 analyses per sample" – This seems to be referring to the data processing steps, though I don't understand why performing these steps multiple times wouldn't produce the same results for each spectrum. And further, I don't understand where the 30 analyses stated here comes from (10 grains * 3 acquisitions per grain * 3 "processes carried out for each spectra" = 90, not 30).

Thank you for highlighting the confusion here. The 3x5sec acquisitions per grain produces one spectrum, which is then deconvolved 3 times. This entire process is carried out for 10 grains per sample. Therefore, 10 grains * 1 spectrum * 3 deconvolutions = 30 results. We have tried to clarify the fact that only 1 spectrum arises from the 3x 5-second acquisitions by adapting the sentence to read, "Each run comprised three co-adds of 5 second acquisitions to produce a single spectrum for analysis. This process was carried out on 10 individual grains from each sample." We have also removed the word "acquired" from line 214 to prevent further confusion.

As for the point about 3 deconvolution repeats, this is to minimise error from the user-guided part of the baseline subtraction process. The user must select pinning points along the spectrum to which the software fits a cubic spline interpolation. Particularly for poorer spectra, the position of the fitted/subtracted baseline can vary significantly depending on the exact pinning point selected. Of course, we could have taken this a step further and involved multiple users in order to minimise bias, but that is perhaps beyond the scope of this study. We have added a note about this in Line… reading "This process was carried out 3 times for each spectrum to minimise the error involved in the user-guided baseline removal process, resulting in 30 analyses per sample."

Line 245 – how is Ro_eq determined from the Raman parameters? (i.e., what equation is used?) It is strange to provide these equations here but still require the reader to go back to Schito and Corrado to see how the Raman parameters come into play. I think

there is no reason to include the equation for T1 here without also presenting the method to determine Ro_eq.

Thank you for highlighting this. We have now included the Schito & Corrado (2018) equation.

Line 281 - "high error causes these ranges overlap" What are the errors? How are these determined? They are not shown on the figure nor in the supplemental data table. Why not plot the error bars associated with each sample on the plots?

We have addressed this point by adding error bars to our figures.

Line 395 – "… or that the equation is not applicable in this instance due to the Raman parameters used"? What is meant by "due to the Raman parameters used"?

This sentence was originally rephrased to read, "This could be due to a weak geothermal gradient, or that the equation contains terms which are affected by strain, therefore altering the results." However, we have since omitted this section for clarity (see earlier comments).

Line 400 – this is difficult for the reader to evaluate because the equation incorporating this parameter is not provided

Addressed by adding the equation – see above response

Line 420 – "significant variation on a sub-km scale" – what, specifically, is meant here? (put a number of the variation; this is relevant for the following discussion)

Thank you for pointing out this imprecision. Our intention was to point out that values exhibited by adjacent samples varied to the extent that a trend was only visible on scales greater than a km, but you are right to point out that a number should be defined with such a statement. However, we have since omitted the section comparing the various geothermometers (see above general comment).

Section 7.8 – The RSCM thermometers investigated in this study are assessed here, based on how much the estimated temperatures differ for the "strained" and "background" samples. For instance, the Kouketsu thermometer is stated to be "more suited to strained environments" because the strained and background samples give similar results (line 431). But I think this misses the point. The goal of this entire process is to, as accurately as possible, estimate the peak temperatures experienced by rocks. A given thermometer equation can yield results that are insensitive to a particular factor (such as strain), but that does not mean that it produces reliable estimates. A key question is, does strain organize organic material in a fashion similar to increasing temperature? If so, I think the goal should be to build a model that connects the nature of the CM (as measured via Raman) to both temperature and strain. It would be ideal if the study had been conducted in a setting where independent estimates of temperature were available (like in the various studies used to calibrate versions of the thermometer). Without this, the authors need to rely on the internal consistency of the results, which can provide interesting insights but can't address the question of which of the applied thermometers gives the most reliable results in the study area.

We appreciate this well thought out comment and agree that an ideal study comparing various geothermometers would indeed include independent temperature estimates. Since we have decided to focus the revised manuscript on the individual parameters only, with greater emphasis on the strained transects, we have omitted the section which compares the various geothermometers (apart from a discursive point on applying our results to the Schito and Corrado equation). This is beneficial both in terms of clarifying the aims of the current paper but also in that it opens up the opportunity to expand the scope of a future study which deals with the comparison of these geothermometers as its principal aim. Such a study would of course include independent temperature estimates which we were unable to obtain for this study due to time, funding, and travel constraints.

Line 432 – "lower than expected for the region…" – on what is this expectation based? In the earlier part of the paper, all that is said about these rocks is that they are "subgreenschist facies" – there don't seem to be any detailed controls on peak temperature here.

Due to various limitations (see above comment) we were unable to obtain independent temperature constraints beyond those gained through simple calculations relating to burial depth and metamorphic facies based on the exiting literature. As it happens, the section referred to in this specific comment has now been omitted (see above).

Line 433: "shows variation in temperature predictions on a sub-km scale, making it less suitable for general use" – I'm not sure what is meant by "general use" here, and I also don't follow the logic. I interpret this to mean that the temperature estimates made using this thermometer are noisy, and therefore unreliable. I worry the authors underestimate the uncertainty inherent in these techniques.

Thank you for pointing out another instance of imprecise language in the manuscript; this is something that we have endeavoured to rectify throughout the revised text. Your interpretation here is correct; however, we are also aware of the "inherent uncertainty" that you mention here. Once again, this geothermometer comparison section has been omitted for the purpose of clarifying the focus of the paper.

Line 435: "… demonstrates a more consistent error…" – what is a "more consistent error"? There are no independent estimates of the temperature for these rocks, so there doesn't seem to be a way to estimate the accuracy of the various thermometers. Do the authors mean that the results for the Lahfid equation for the strained samples are consistently biased in the same direction? I think that is different than having a "consistent error", which I would take to reflect the magnitude of a temperature difference from a true value.

Once again, thank you for highlighting our choice of language – and again, your interpretation is correct. We refer to a consistent shift direction, and although the Lahfid equation is no longer used, we have ensured that this terminology is used in the rest of the paper (e.g. Line 369).

Line 435: "…the predicted temperatures are more in line with those predicted for the area…" Again, based on what? There don't seem to be any independent temperature estimates.

This is based on burial depth – however, we no longer compare multiple geothermometers in this work (see earlier comments).

Line 460: "… an equation is required that can resolve temperature changes over hundreds of metres at the least" – is this even feasible? There are errors in all of these measurements.

This is an excellent question, and not one we can even attempt to answer within the scope of this study! However, future work and the continued development of Raman geothermometric techniques will hopefully provide an answer.

Line 461: "we conclude… choosing the most appropriate equation is complex and dependent on multiple factors" – what are the "multiple factors"? The preceding discussion suggests that strain is one, but I'm not sure what else is meant here.

To clarify this, we now refer only to strain in the concluding remarks, and as mentioned above no longer compare multiple equations. We do however mention that "the fact that not all strained samples produce calculated temperature shifts of the same direction or magnitude suggests that the process is more complex than simply strain or temperature having an effect" (Line 507)

Line 465: "The use of multiple parameters… suggests that the equation should be relatively insensitive to strain" – I don't understand this. If any of the parameters that go into the temperature estimate are influenced by strain, why wouldn't that influence the results? Why would including other parameters limit the influence of parameters sensitive to strain?

The reasoning behind this statement follows the idea that an equation which is based on one parameter only will be 100% affected by that parameter's sensitivity to strain. If multiple parameters are used in an equation, and not all of them are altered by the introduction of strain, then the result of the equation will not still be affected but not to the same degree.

Line 468: "The use of multiple terms in the equation may help to produce more reliable results (as the influence of different parameters interact)" – why? The logic here is not clear to me. I think what matters is the inclusion and appropriate weighting of the relevant parameters, not the number of terms in the equation.

Our point here refers to the weighting of parameters, as you suggest, but our wording was not clear. As we no longer compare multiple geothermometers, we do not discuss the relative benefits of weighting certain parameters, only alluding to this point in Line 505 where we state "The most significant term in the equation is I[d]/I[g], and our data shows that I[d]/I[g] is strongly affected by strain-related spectral changes. It therefore follows that the equation should be sensitive to strain…"

Technical corrections

Line 21 – should be "up to 140"

Since we no longer analyse multiple geothermometers we have omitted this statement from the abstract.

Line 100 – grammar (replace semi-colon with " and…")

Changed – thank you.

Line 171 – Reword; the text here implies that Raman spectroscopy works only on organic carbon.

Text now reads "Raman spectroscopy measures the wavelengths of radiation produced by inelastic (Raman) scattering during the de-excitation of electrons in different molecular bonds, in this case focussing on those involved in different forms of organic carbon." (Line 222).

Line 178 – Is "pertaining" the right word here?

Perhaps not – this has been changed to "approaching" (Line 233)

Line 261 – should be "detail", not "detailed"

Changed – thank you.

Line 277 – units should be included with these values

We appreciate that unitless values look strange, but as we are presenting ratios we cannot give them units.

Figure 4 – why are some of the grey lines dashed rather than solid? There is no explanation for this on the figure or in the caption. Other than this and the lack on uncertainties shown on the data, the figures are very attractive and quite helpful, especially the maps and cross sections (good work!).

Thank you for pointing out our failure to explain this. The dashed lines are to indicate where trends are not directly indicated in the literature but are inferred from related parameters.