Reviewer 2:
Manuscript Number: bg-2022-207

General Comments

This manuscript and the associated data provide a high value set of SOC fraction gridded products for Australia and will be an important resource for land managers and the scientific community. There are some question marks over the reliability of the data due to bias in the original calibration samples (primarily agricultural soils) and in the method used to derive the PyOC and MAOC fractions for the calibration samples. The approach used can clearly be made more robust in future by starting with a larger, more representative and more reliable set of calibration data.

Addressing the review criteria:

1. Does the paper address relevant scientific questions within the scope of BG?
   Yes
2. Does the paper present novel concepts, ideas, tools, or data?
   Yes. The publication and data will be of considerable use to management and science communities in Australia and globally.
3. Are substantial conclusions reached?
   Yes.
4. Are the scientific methods and assumptions valid and clearly outlined?
   This are some weaknesses in the scientific methods/sampling that are the basis of this manuscript. The authors point out issues in the sampling, where there was insufficient range of soil types/biomes used in the initial calibration. What they do not point out is that the method used to estimate the fractions is not particularly reliable.

   The SOC fractions used for calibration of the spectral methods were measured using a chemical approach (and data) developed for SCaRP a decade ago. The ^{13}C NMR based approach to determine PyOC is not widely used for estimating concentrations/stocks as it is a semi-quantitative method (e.g it often under-detects aromatic C and spectral assignment /integration is difficult in low-SOC samples).
   The current global data set of PyOC (black carbon) has been obtained using the Benzene PolyCarboxylic Acid (BPCA) approach, this approach gives a more realistic estimate of concentrations of PyOC in soil (see Jones, A. et al. "Fires prime terrestrial organic carbon for riverine export to the global oceans," Nat. Commun. 11, 2791 (2020). https://doi.org/10.1038/s41467-020–16576-z', Dymov, A.A. et al. Comparison of the Methods for Determining Pyrogenically Modified Carbon Compounds. *Eurasian Soil Sc.* **54**, 1668–1680 (2021). https://doi.org/10.1134/S1064229321110065)
5. Are the results sufficient to support the interpretations and conclusions?
   Yes.
6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?
   Yes.
7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution?
   Yes.

8. Does the title clearly reflect the contents of the paper?
   Yes.
9. Does the abstract provide a concise and complete summary?
   Yes.
10. Is the overall presentation well structured and clear?
    Yes.
11. Is the language fluent and precise?
    Yes.
12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?
    Yes.
13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?
    No.
14. Are the number and quality of references appropriate?
    Yes.
15. Is the amount and quality of supplementary material appropriate?
    Yes.

Specific Comments

2 Materials and Methods

1. PyOC was estimated by ^{13}C CP MAS NMR, this is excellent for identifying the types of carbon present but is generally regarded as only semi-quantitative in nature. The data for the fractions have not been cross-correlated with other approaches such as BPCA to ensure that they are in fact robust. MAOC is estimated by difference subtracting POC and PyOC from SOC, as a result there is also a question mark over the reliability of the estimates of this variable.
What is needed are measures of the error associated with PyOC and MAOC and how this transfers to errors in the subsequent estimates of these fractions using the irl1 and irl2 models.

2. The process of generating MAOC, POC and PyOC is reliant on an initial calibration and this was done on 312 samples a decade ago. A much larger number of samples were used in the spectral harmonisation and data set modelling but in the end they are reliant on this small number of samples (312) for SCaRP and an even smaller number of SCaRP samples (200) for the AusSpecMIR and AusSpecMIR2 and 309 for AusSpecIR. Again the authors need to justify how the use of this small set of samples, dominated by agricultural soil types, is able to successfully be used to provide calibration data for the much wider set of biome/soil types used in this study.

This may help to explain why the authors experienced difficulty with their preferred approach of modelling SOC fraction concentrations directly: '*The Pearson's r correlation coefficient 220 was 0.56, but the sum of SOC fractions showed some extreme values (Figure S1).*

3. The authors have used a very thorough and well thought through approach to generate a spatially and depth consistent gridded set of SOC fraction data for the continent.
Figure 2 (I think) shows where the spectral training data sets were located. It would be useful to see where the original SCaRP SOC fraction calibration data was collected, either on a map of Australia or in a table by soil/biome type, this would provide the reader with a clearer idea of the limitations due to type of calibration data used.

3 Results

4. The reporting of errors is an issue in the manuscript.

The authors need to do a major check over all sections of the text and tables to ensure that precision is treated correctly. Errors should have 1 or at most 2 significant digits.
The problems start in the abstract, *59% ±17.5%, whereas 28% ± 17.5% was PyOC and 13% ± 11.1%* in this case the errors have more decimal places than the values and there are too many significant digits for such large errors.
The estimate of stocks *12.7 Pg MAOC, 2 Pg POC* has inconsistent precision, possibly this is correct but given the other issues possibly not.
As a clearer example the authors report (L388) *13.1% ± 11.1%,* this should be 13 +/- 11 which indicates 85% error, reporting to +/- 0.1 (0.7%) clearly makes little sense.
The decimal place in the error sets the decimal place in the value, they should always agree.
Things get worse : Table 3. *2.49 +/- 118.3*   *0.64 +/- 23.8*

5. It would be very useful to have stocks with errors estimated based on the data generated from the grid. The authors estimate stocks but then they provide no errors because of an issue around soil thickness. Without an error then these stocks are of limited use (see above).
It would be better to have some estimates of error for these stocks that sum over the best estimates for soil thickness, issues around soil bulk density estimates etc.

4 Discussion

6. Mention is made of a likely underestimate of SOC in forest systems and it is clear the authors were aware of the lack of calibration sampling in forest systems. *'or the fact that the fractionation in the original dataset was applied to agricultural soils and some pastures but lack forest soils.' 'The uncertainty on the spatial predictions of SOC fraction stocks was driven mainly by TOC and the proportion of SOC fractions predictions, which in turn rely on spectral predictive models developed with soil samples originating mainly from agricultural soils.'*
Australia has 134 million hectares of forest, 17% of the land surface area.
https://www.agriculture.gov.au/abares/forestsaustralia/australias-forests#forest-area
It is somewhat surprising that they did not add additional calibration samples from the forest estate into the early stages of this study. The potential magnitude of this underestimation might be mentioned.

Technical Comments

Line 73
In Australia, the long history of burning suggests pyrogenic organic carbon (PyOC) is an additional important component (Lehmann et al., 2008) of SOC. PyOC refers to charred residues derived from

Line 75
comprised by a continuum of organic compounds thermally altered by fire

Line 90
the contribution of SOC fractions (MAOC, POC, and PyOC) to the total SOC in the top 30 cm of the soil and update the Soil

Line 101
mapped proportions of SOC. Finally, we calculated SOC fraction stocks for the 0-30 cm topsoil using data from SLGA maps

Line 135
The content of poly-aryl C

Line 164
resolution. Soil samples from AusSpecMIR and AusSpecMIR2 were scanned in quadruplicate.

Line 180
soil), total organic carbon (TOC) concentration (mg C g^{-1} soil), and used harmonised spectra.

Line 241
The Soil Data Federator is a web API that compiles soil data from different institutions

Line 292
(Meinshausen, 2006). In Digital Soil Mapping (DSM), quantile regression

Line 445
for ilr2 the importance of gamma radiometrics variables and gravity was more important for the