General comments: This manuscript evaluated Vis–NIR spectroscopy for laboratory and in-situ prediction of forest soil organic carbon fractions. Authors found that the highest accuracy for POC was achieved using EPO-corrected in-situ spectra (R^2 = 0.90), whereas MAOC prediction performed best using uncorrected in-situ spectra (R^2 = 0.71). The investigated topic is interesting for the forest ecosystem and this manuscript is generally well-written with clear objective, solid methodology and insightful discussion. However, several limitations should be addressed before acceptance: (1) authors should provide more deep discussion on the better model performance of POC than MAOC, which is in contract to most of current studies; (2) authors can provide more information on the performance difference among topsoil and subsoil since the subsoil is getting more attention due to limited understanding. (3) please carefully check the formatting in the whole manuscript.

General response: We thank the reviewer for the positive and constructive assessment of our manuscript. We have carefully addressed all three comments. In particular, we have expanded the discussion to better explain the higher prediction performance of POC relative to MAOC, further investigated and discussed differences in prediction performance between topsoil and subsoil, and conducted a comprehensive check of the entire manuscript to correct formatting, citation, and consistency issues. Detailed responses to each specific comment and the corresponding revisions are provided in the following sections.

Specific comments:

1. **Line 22: It is not clear the difference between EPO-assisted transfer and EPO-assisted in-situ modelling.**
2. **Line 24: In contrast, direct in-situ modeling, this sentence is not complete.**

   Response: Thank you for pointing this out. The original wording did not clearly distinguish the four in-situ application workflows, and the phrase "direct in-situ modeling" in Line 24 resulted in an incomplete sentence. We have therefore revised this part of the Abstract to explicitly describe each workflow according to the model calibration domain and the type of in-situ spectra used. The revised sentence in the Abstract now reads:

   '*This study developed an integrated framework to evaluate in-situ Vis–NIR spectroscopy for predicting SOC fractions by comparing four application workflows: (A) direct application of laboratory-calibrated models to raw in-situ spectra; (B) application of laboratory-calibrated models to external parameter orthogonalization (EPO)-corrected in-situ spectra; (C) direct modelling using raw in-situ spectra; and (D) modelling using EPO-corrected in-situ spectra.*'

   This revision makes it clear that Workflow B involves transferring a laboratory-calibrated model to EPO-corrected in-situ spectra. In contrast, Workflow D involves model development using EPO-corrected in-situ spectra. We have also revised the incomplete sentence associated with "direct in-situ modeling" accordingly.

3. **Lines 28-29: Generally, MAOC is associated to clay minerals, while the finding here shows that POC is linked to organo–clay absorption features. Any explanation for this?**

   Response: We appreciate the reviewer helping us clarify this point, which highlights an important nuance in spectral interpretation. We would like to clarify that our finding does not imply a direct chemical binding

between POC and clay minerals. Instead, it reflects a composite spectral response arising from overlapping absorption features and physical aggregation in these soils, as explained below.

The spectral region around 2200 nm contains overlapping absorption features from both organic functional groups (C–H combination overtones and phenolic O–H groups) and clay mineral lattice vibrations (Al–OH bending/stretching modes). In our coarse-textured albic forest soils, POC dominates the SOC pool (>80% of SOC, mean 19.04 g kg^{-1}), existing predominantly as decomposing plant debris within soil macro-aggregates (>53 μm). Within these macro-aggregates, particulate organic residues are physically entangled with fine clay coatings. Therefore, the strong contribution around 2200 nm to POC prediction represents a composite spectral response caused by the spectral overlap and spatial covariance between organic C–H/O–H overtones and background clay Al–OH vibrations, rather than a direct molecular-level organo-clay chemical bond.

Although MAOC is chemically associated with clay surfaces (<53 μm), Vis–NIR spectroscopy (400–2500 nm) lacks the primary fundamental vibrational modes—such as microbe-derived amide I–II bands (~ 1660–1550 cm^{-1}) and metal oxide lattice features—that directly resolve MAOC under MIR spectroscopy. Consequently, Vis–NIR models cannot directly detect the MAOC molecule itself; instead, they rely on indirect mineral-mediated proxies (such as visible-range iron oxide color signatures and surface background scattering) as statistical surrogates. In our albic soils, where podzolization eluviates free iron oxides and fine clay, these indirect mineral proxies are attenuated, leading to weaker direct spectral expression for MAOC compared to the strong organic SWIR overtones of POC. To prevent any potential misunderstanding, we have rephrased the sentence in the Abstract as follows:

'*Spectral interpretability analysis demonstrated that POC prediction was driven primarily by organic C–H/O–H combination overtones in the shortwave infrared region (2150–2300 nm, encompassing composite organic–clay spectral overlap around 2200 nm), whereas MAOC prediction was indirectly mediated through visible-range mineral proxy signatures.*'

Furthermore, we expanded the mechanistic discussion in Section 4.3 to thoroughly explain this composite overlap: *'The contribution around 2200 nm, however, should be interpreted cautiously because this region contains overlapping absorption features associated with both organic constituents and clay-mineral Al–OH vibrations. Thus, its strong contribution to POC prediction does not necessarily indicate a direct POC–clay association, but may instead reflect a composite spectral response arising from covariance between organic and mineral-related spectral variation in these soils.'*

4. **Line 59: Please make the citations consistent (R. A. Viscarra Rossel et al., 2006; Rossel and Behrens, 2010).**

   Response: Corrected. These citations have been standardized to (Viscarra Rossel et al., 2006, 2016) in the manuscript.

5. **Lines 64-65: Indeed, there are much more studies on prediction SOC fractions using vis-NIR spectroscopy. Please see some examples below and provide a comprehensive review on previous work.**

Viscarra Rossel, R. A., and W. S. Hicks. "Soil organic carbon and its fractions estimated by visible–near infrared transfer functions." European Journal of Soil Science 66.3 (2015): 438-450.

Cayuela-Sánchez, José A., and Rafael López-Núñez. "Compositional Data Methods and VISNIRS to Predict Soil Organic Carbon Contents." European Journal of Soil Science 76.5 (2025): e70200.

O'Rourke, S. M., and N. M. Holden. "Determination of soil organic matter and carbon fractions in forest top soils using spectral data acquired from visible–near infrared hyperspectral images." Soil Science Society of America Journal 76.2 (2012): 586-596.

Response: We thank the reviewer for suggesting these valuable works. We have revised paragraph 3 in the Introduction; specifically, it now reads as follows:

'Diffuse reflectance spectroscopy in the visible–near infrared range (Vis–NIR, 350 to 2,500 nm) provides a promising alternative for rapid soil analysis. Because soil organic matter and minerals exhibit characteristic absorption features across the Vis–NIR region, this technique enables the simultaneous estimation of multiple soil properties (Viscarra Rossel et al., 2006, 2016). Over the past two decades, laboratory-based Vis–NIR spectroscopy has demonstrated strong predictive capability for bulk SOC and other soil properties across various ecosystems (Viscarra Rossel et al., 2006; Lu et al., 2013; Dematt̂e et al., 2017; Jaconi et al., 2017; Bai et al., 2023). Recent methodological advances have further enhanced its utility, for instance, by applying hyperspectral imaging to evaluate organic matter in forest topsoil (O'Rourke and Holden, 2012) or integrating compositional data (CoDa) analysis to mitigate soil moisture interference in Vis–NIR-based SOC predictions (Cayuela-Sánchez and López-Núñez, 2025). Despite these advances, the application of Vis–NIR spectroscopy to SOC fractions remains comparatively limited. Some studies have explored the use of Vis–NIR spectroscopy to estimate operational carbon pools, such as humic and resistant organic carbon (Viscarra Rossel and Hicks, 2015). By comparison, spectroscopic investigations of SOC fractions have predominantly relied on mid-infrared (MIR) spectroscopy, which provides access to fundamental vibrational features of organic functional groups and mineral matrices (Janik et al., 2007; Pacini et al., 2025). Consequently, relatively few studies have explored the potential of Vis–NIR spectroscopy to predict functionally distinct POC and MAOC (Qi et al., 2024), particularly under in-situ conditions.'

6. **Lines 130-131: Were laboratory spectra measured on the dried soil samples sieved through a 0.15-mm mesh?**

Response: Thank you for this clarifying question. We would like to clarify that laboratory Vis–NIR spectra were actually acquired on air-dried soil samples sieved through a 2-mm mesh (as mentioned in Section 2.2), rather than a 0.15mm mesh. The 0.15 mm sieving step was used exclusively for elemental carbon analysis of the separated organic carbon fractions (Section 2.2). To prevent any confusion and ensure clarity, we have explicitly specified the sample preparation details for spectral acquisition in Section 2.3 as follows:

'Laboratory spectra were obtained from air-dried, ground, and sieved samples (sieved through 2-mm mesh) placed in 8-cm diameter containers with leveled surfaces.'

7. **Line 164: Better to mention Location-based KS algorithms in the flowchart.**

Response: Due to spatial layout constraints within the flowchart diagram boxes, adding lengthy text directly into the graphic box would cause visual clutter. Therefore, we have incorporated a detailed, explicit description of the Location-based KS algorithm directly into the caption of Figure 2 as well as in Section 2.4.1. Specifically, we added the following detailed explanation to the Figure 2 caption:

'The train and test sets were partitioned using spectral-based Kennard–Stone (KS) sampling with location-level grouping.'

And section 2.4.1:

'T*he laboratory spectra were partitioned into calibration and test sets using the Kennard–Stone (KS) algorithm (Kennard and Stone, 1969) based on spectral distances to ensure representative coverage of the laboratory spectral space.*'

8. **Line 257: RPIQ would be more robust than RPD.**

Response: RPIQ is indeed more robust for skewed distributions than RPD. We have added the formula for RPIQ to Section 2.5 (Eq. 5), but we also save RPD as indicator results value, report RPIQ in some places in the manuscript text and figures, and update all model accuracy results in the supplement Table S2.

9. **Lines 294-300: It would be more informative to present the difference among different depth intervals.**

Response: We thank the reviewer for this constructive suggestion. In response, we have expanded Section 3.1, Section 3.2, and Supplementary Table S1 to detail the depth-stratified distributions of POC and MAOC, fraction stoichiometry, physical texture, and spectral features. Specifically, both fractions exhibit a marked downward concentration gradient accompanied by a contraction in spatial variability, while the relative contribution of MAOC increases progressively in deeper horizons. Furthermore, we expanded Section 4.4 to discuss the trade-offs of profile-wide unified modeling versus depth-stratified modeling, offering perspectives for future subsoil carbon monitoring.

The revised text in Section 3.1 now reads:

'Furthermore, both POC and MAOC concentrations displayed pronounced downward gradients across the four soil depth intervals (0–10, 10–20, 20–30, and 30–40 cm), with mean concentrations declining from 52.64 to 4.49 g kg^{-1} for POC and from 7.71 to 2.44 g kg^{-1} for MAOC, respectively, accompanied by a gradual increase in the relative contribution of MAOC to total measured SOC. Across the profile, the POC: MAOC ratio gradually shifted from 6.8:1 in the topsoil (0–10 cm) to 1.8:1 in the subsoil (30–40 cm), accompanied by a doubling of clay content from 7.5% to 16.1% (Table S1).'

Additionally, to address the broader implications for subsoil modeling, we expanded the Discussion in Section 4.4 with the following perspective on depth-dependent observability and future prospects:

'Topsoils (0–10 cm) exhibit exceptionally high POC accumulation (mean >50 g kg^{-1}) and a broad dynamic

range, yielding a higher spectral signal-to-noise ratio where moisture correction (EPO) exerts its greatest efficacy. Conversely, subsoil layers (10–40 cm) show a sharp drop in POC (declining to <5 g kg⁻¹) alongside a doubling of clay content (from 7.5% to >16%), where organic functional group signals become attenuated relative to mineral light scattering. As larger, depth-dedicated regional field datasets are compiled in the future, horizon-stratified modeling coupled with depth-resolved spectral feature extraction will represent a valuable avenue to further refine in-situ predictions of subsoil carbon fractions.'

10. **Lines 459-501: Based on a recent review by Ding et al. (2026), MAOC can be better predicted than POC, which is against the findings in this work. Please provide deep discussion on this point.**

Ding, S., Xu, Y., Agathokleous, E., Li, T., Zheng, H., & Feng, Z. (2026). A global synthesis of spectroscopy-based prediction accuracy for soil carbon fractions: A systematic review. Soil and Tillage Research, 262, 107242.

Response: We thank the reviewer for highlighting this important point. By integrating the regional empirical findings of Dai et al. (2024) and the global synthesis conclusions of Ding et al. (2026), we conducted a multi-level comparative analysis to address this apparent divergence in our revised manuscript (Section 4.1 and Section 4.3). Specifically, this divergence from the general pattern observed in previous cropland-dominated syntheses is driven by three interconnected mechanisms:

1. Distinct Pedogenic Context and Fraction Stoichiometry:

   The regional study by Dai et al. (2024) analyzed agricultural croplands (e.g., wheat–rice rotation fields on Alisols or Fluvisols), where MAOC dominated the SOC pool (averaging ~76% of SOC, with 14.6 g kg^{-1} MAOC vs. 5.5 g kg^{-1} POC), consistent with the cropland-dominated context represented in the global synthesis of Ding et al. (2026). In contrast, our study investigated forest soils characterized by albic and podzolization-related features, where POC dominated the SOC pool and accounted for more than 80% of total measured SOC (mean POC = 19.04 g kg^{-1} vs. MAOC = 4.56 g kg^{-1}; POC: MAOC ≈ 4:1). The soils were predominantly silt-rich, with approximately 74% silt, 13% sand, and 14% clay on average across the profile (Table S1). Strong eluviation associated with podzolization may reduce reactive Fe/Al phases and the availability of fine mineral surfaces in upper horizons, potentially weakening mineral-related spectral features relevant to MAOC prediction. Meanwhile, substantial forest litter inputs sustain a large POC pool. These contrasting organic-matter input regimes and mineralogical conditions may therefore contribute to the different spectral expression and prediction performance of POC and MAOC in our study. This interpretation is consistent with Ding et al. (2026), who identified soil texture, mineralogical context, and target-variable dynamic range as important factors influencing the prediction accuracy of SOC fractions, particularly MAOC.

2. Concentration Dynamic Range and Spectral Signal-to-Noise Ratio:

   POC exhibited a broad concentration range (1.76–67.89 g kg^{-1}; up to 176 g kg^{-1} in topsoil) and substantial variance across samples. This broad dynamic range provides a wide spectral-reference gradient and high

signal-to-noise ratio, enhancing statistical detectability during calibration. Conversely, MAOC had a much lower abundance (mean 4.56 g kg^{-1}) and a narrow reference distribution (0.48–17.62 g kg^{-1}), which restricts the range of spectral-reference covariance and constrains prediction accuracy.

3. Spectroscopic Region Mismatch (Vis–NIR vs. MIR):

In their synthesis, Ding et al. (2026) reported that MAOC prediction achieves significantly higher accuracy under mid-infrared (MIR) spectroscopy than under near-infrared (NIR) spectroscopy ($r \approx 0.91$ for MIR vs. 0.84 for NIR, $P < 0.01$; where the NIR category included Vis–NIR studies). Vis–NIR spectroscopy (400–2500 nm) effectively captures POC through direct SWIR C–H and O–H combination overtones (2150–2300 nm). However, Vis–NIR lacks the primary fundamental vibrational modes—such as microbe-derived amide I–II bands (~1660–1550 cm^{-1}) and metal oxide lattice features—that dominate MIR MAOC detection. In mineral-depleted albic horizons, Vis–NIR prediction of MAOC experiences an inherent spectroscopic sensitivity threshold.

The specific text in Section 4.1:

The relative difference in prediction accuracy between POC and MAOC may partly reflect the distinctive pedogenic and mineralogical characteristics of the forest soils investigated here. Unlike the low-altitude agricultural systems examined in previous studies (e.g., wheat–rice rotations on Alisols or Fluvisols derived from lacustrine and marine deposits, where MAOC dominated the SOC pool at an average of 76% [14.6 g kg^{-1} MAOC vs. 5.5 g kg^{-1} POC out of 20.0 g kg^{-1} SOC]; Dai et al., 2024), our soils developed under a forest environment characterized by albic/podzolization-related features (Albic Luvisols or Podzols). In our forest soils, POC dominated the SOC pool, accounting for more than 80% of total measured SOC (mean POC of 19.04 g kg^{-1} vs. MAOC of 4.56 g kg^{-1}; Fig. 3). The soils in the present study were dominated by silt-rich textures, comprising approximately 74% silt, 13% sand, and 14% clay on average across the profile (Table S1). In such albic forest soils, prolonged eluviation associated with podzolization severely reduces the abundance of fine clay and reactive Fe/Al mineral complexes in upper horizons, directly attenuating the diagnostic mineral absorption signatures (Fe–O charge transfer < 550 nm and Al–OH lattice vibrations at 2200 nm) that normally drive MAOC quantification (Hartemink et al., 2020; Lundström et al., 2000). At the same time, heavy inputs of forest litter sustain a large POC pool. Together, these pedogenic conditions alter the balance between particulate and mineral-associated carbon by modulating both organic matter input regimes and the availability of reactive mineral surfaces for organo-mineral association (Kleber et al., 2015). Therefore, the POC-dominated SOC composition and its superior predictive performance ($R^2 = 0.96$) reflect the combined influence of forest organic inputs and quartz-rich albic pedogenesis, aligning cleanly with the global synthesis of Ding et al. (2026), which identified soil texture, mineralogical context, and concentration dynamic ranges as dominant environmental drivers governing SOC-fraction prediction accuracy, particularly for MAOC.'

And Text of Section 4.3:

This indirect and context-dependent spectral representation may help explain the comparatively lower prediction accuracy of MAOC observed in this study. This interpretation is also consistent with the global

synthesis of Ding et al. (2026), which reported that NIR spectroscopy can capture variation in POC through SWIR overtone and combination absorptions, whereas MAOC showed stronger prediction performance with MIR spectroscopy, where fundamental vibrational features of organic functional groups and mineral matrices are more directly resolved.'

11. **Some references are not complete and correctly cited, such as Dai, L., 2025 (the journal information is missing), Rossel, R.A.V. (should be Viscarra Rossel, R.A.), Behrens, T., 2010.**

    Response: Thanks to the reviewer's careful check. We have audited the entire reference list and corrected all formatting errors.