Comment on Hodgkins et al. (2018)

Henning Teickner[1,*], Klaus-Holger Knorr[1]

[1] ILÖK, Ecohydrology and Biogeochemistry Group, University of Münster, Heisenbergstr. 2, 48149 Münster, Germany

[*] Corresponding author: Henning Teickner (henning.teickner@uni-muenster.de)

Introduction

Hodgkins et al. (2018) used mid infrared spectra (MIRS) to make inferences about the stability of peat against decomposition along a latitudinal gradient from the tropics to northern latitudes. Recently, we have shown that these spectral prediction models are biased and their predictions more uncertain than considered in Hodgkins et al. (2018; Teickner and Knorr 2022). Here, we show what consequences this bias and uncertainty and additional neglected uncertainty sources have for the main conclusions in Hodgkins et al. (2018).

In particular, we find that:

1. Larger aromatic contents may be necessary to stabilize tropical near-surface peat against decomposition and aromatics may accumulate at a higher rate with depth in tropical peatlands than estimated by Hodgkins et al. (2018).

2. Larger uncertainties indicate that also larger differences in aromatic contents between (sub)tropical and high latitude peat than estimated by Hodgkins et al. (2018) are possible, also between deeper peat.

3. More research should focus on how estimates of carbohydrate and aromatic contents from MIRS may be confounded by other organic matter fractions, in particular proteins. As a first step, this requires accurate concepts to name organic matter fractions and variables used in the interpretation of MIRS.

These uncertainties have the potential to change predictions of models on global peat stability as well as what stabilizes peat against decomposition if temperatures rise and thus add in particular to the debate to what extent global warming may increase decomposition of northern deep peat deposits (e.g. Dorrepaal et al. (2009), Wilson et al. (2016), Baysinger et al. (2022)).

That said, theoretical considerations of decomposition processes alone support the suggested gradient in peat chemistry and Hodgkins et al. (2018) made an important contribution in

providing the first open access models and estimates for peat carbohydrate and aromatics content on a large scale. Increasing the accuracy of models is an important part of the scientific process and we hope that this comment will focus research on the problems which hamper improving this accuracy.

In the following, we will use the terms holocellulose instead of the more general term carbohydrates and Klason lignin instead of the more general term aromatics. These are the accurate terms for the organic matter fractions quantified in Hodgkins et al. (2018).

Uncertainty sources not considered in Hodgkins et al. (2018)

In a recent study, we have shown that the models used to estimate holocellulose and Klason lignin contents in Hodgkins et al. (2018) (original models) are not valid for peat and this will cause larger uncertainties and biased predictions (uncertainty source 1) — especially for decomposed peat — of the estimated holocellulose and Klason lignin contents (Teickner and Knorr 2022).

Besides this, the following two uncertainty sources have not been considered in Hodgkins et al. (2018): first, the prediction uncertainty of the spectral prediction models (uncertainty source 2), and second, the uncertainty introduced when computing near-surface average holocellulose and Klason lignin contents from estimates for individual peat layers (uncertainty source 3).

We recomputed the models for the latitudinal gradients of near-surface peat holocellulose and Klason lignin contents while considering all three uncertainty sources (supporting information S1).

Since we do not have yet models to accurately predict peat holocellulose and Klason lignin contents from MIRS, it is of course difficult to quantify the uncertainty and bias introduced

by uncertainty source 1. However, the modified models provided in Teickner and Knorr (2022) are very likely more accurate and therefore the difference between predictions of the original model and our modified models are a plausible approximation of this additional uncertainty and bias.

The reanalysis shows that both the estimated slopes for the latitudinal gradient (95% confidence intervals are [-0.1,1.4] and [-1.7,0.1] for holocellulose and Klason lignin, respectively; figure 1 and supporting figure 3) and differences in the depth profiles are highly uncertain due to these uncertainty sources (figure 2).

Thus, our reanalysis shows that estimates of the latitudinal gradient and depth gradients are much more uncertain than previously assumed.

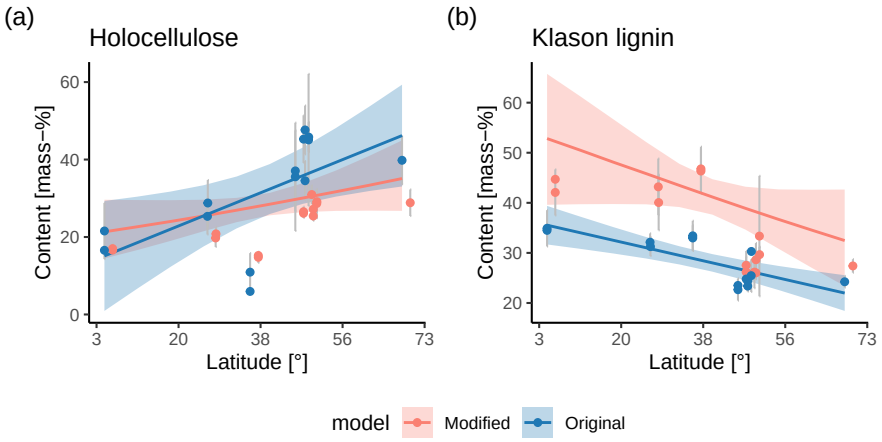

Figure 1: Predicted average surface ($\leq$ 50 cm) peat holocellulose (a) and Klason lignin (b) contents plotted against latitude (compare with Fig. 3 in Hodgkins et al. (2018)). Lines and shaded areas represent average predictions from regression models and 95% confidence intervals. "Modified" is the modified Bayesian hierarchical regression model which simultaneously models individual samples' contents from mid infrared spectra and the latitudinal gradient of average core near-surface peat contents. This model considers prediction uncertainty from the mid infrared spectra and from computing per-core averages. "Original" is the original linear regression model (Hodgkins et al. 2018) computing only the latitudinal gradient of average core near-surface contents. This model does not consider prediction uncertainty from mid infrared spectra, nor uncertainty from computing per-core averages. Points are average core near-surface contents predicted from the model ("Modified") or computed from the average predictions for individual samples ("Original") with error bars representing 95% confidence intervals. Points for "Modified" are shifted by +0.1°.

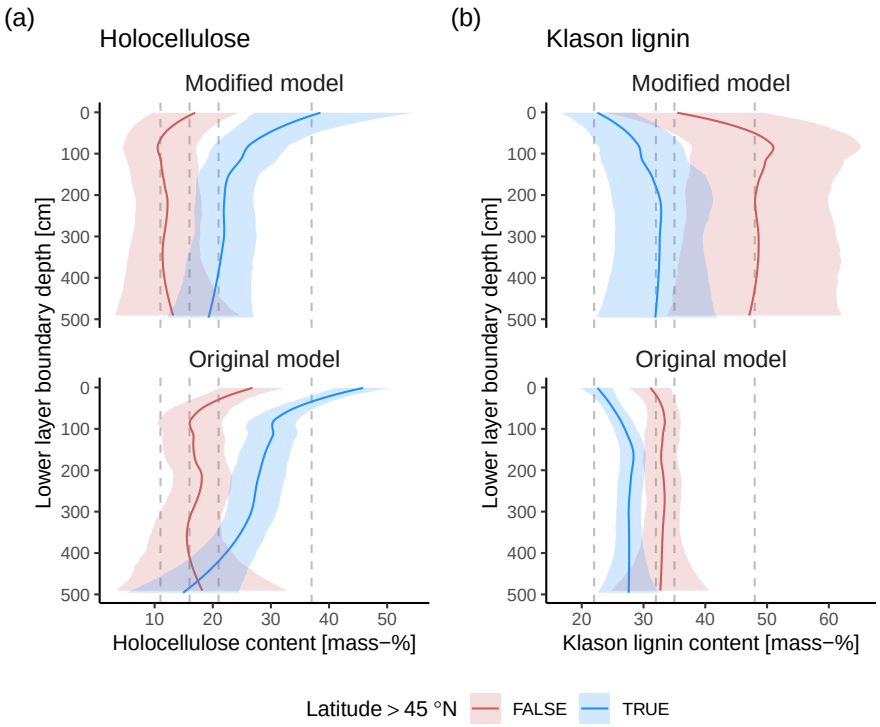

Figure 2: Predicted median holocellulose (A) and Klason lignin (B) depth profiles of peat core data classified into two latitude categories following Hodgkins et al. (2018) (compare with Fig. 2 in Hodgkins et al. (2018)). Lines are averages of LOESS smoothers fitted to the predicted values by the models. Shaded regions are corresponding 95% confidence intervals, comprising prediction uncertainty in the holocellulose and Klason lignin estimates, respectively. Vertical dashed lines in each columns represent approximate surface and average mean contents as predicted using the improved models ("Modified").

Larger aromatic contents may be necessary to stabilize tropical near-surface peat against decomposition

Our previous study has shown that the spectral prediction model for Klason lignin is biased, especially for more decomposed peat (Teickner and Knorr 2022). Our reanalysis using the modified model for Klason lignin from Teickner and Knorr (2022) indicates that larger Klason lignin contents may be necessary to stabilize tropical near-surface peat against decomposition as well as that changes in Klason lignin with depth may be more pronounced in the tropics than estimated in Hodgkins et al. (2018) (supporting information S1).

With our modified model, near-surface peat Klason lignin contents are on average ~10 to 15 mass-% larger across the latitudinal gradient (figure 1). Specifically, average (sub)tropical ($< 45°$N) near-surface peat Klason lignin contents are 10 [-2,26] mass-% (median, lower and upper 95% prediction interval limit) larger than with the original model. The large uncertainties now made explicit would also allow on average larger differences between deep (sub)tropical ($< 45°$N) and high-latitude ($\geq 45°$N) peat (figure 2).

Similarly, residual enrichment of Klason lignin during decomposition may have been underestimated. A rough estimate for the residual enrichment of Klason lignin during decomposition is the difference in Klason lignin content between near-surface peat and deeper peat. With the modified model, this difference is on average for (sub)tropical peatlands 5 [0,9] mass-% larger (figure 2). For high latitude peatlands the difference is smaller and much more uncertain than previously stated (2 [-2,6] mass-%).

Consequently, in general — and especially in (sub)tropical peatlands — the average residual enrichment of Klason lignin due to decomposition probably has been underestimated by the original model. A consequence of this is that high latitude peat deposits may experience more decomposition under a warmer climate than suggested in Hodgkins et al. (2018) because a larger content of Klason lignin is necessary to stabilize peat chemically under warmer conditions.

The need to use precise concepts for organic matter fractions

We argue that we should differentiate between vague concepts such as carbohydrates and aromatics and precise concepts such as holocellulose and Klason lignin and we should always use the most precise concept possible to describe the organic matter fraction we *plan* (or intent) to measure, unless more general statements are explicitly warranted.

For example, Hodgkins et al. (2018) planned to measure Klason lignin contents because their spectral prediction model used Klason lignin data as dependent variable, however this variable is labeled as aromatics. Limitations of the extraction procedure by which Klason lignin are defined are known (e.g., Hatfield and Fukushima (2005), Bunzel, Schüßler, and Tchetseubu Saha (2011), Abu-Omar et al. (2021)) and these limitations as well as differences to other procedures get obscured by using vague words such as carbohydrates and aromatics.

However, using precise concepts for organic matter fractions is also important for exactly the opposite reason: to make clear how much accuracy and precision a variable actually has, instead of implying that it would quantify a variable more accurate and precise than is actually the case. This becomes especially important when interpreting peat chemistry based on mid infrared spectra (MIRS) or spectral prediction models because all variables derived from MIRS — for example the widely used humification indices, as proxy for relative contents of recalcitrant organic matter fractions (e.g. Broder et al. (2012)), or Klason lignin contents predicted from models — are in fact only indirect estimates of organic matter fractions. These indirect estimates of organic matter fractions can be misleading in abundant ways if they are not sufficiently validated. Stating these limitations requires precise concepts.

Intensities recorded in MIRS really only represent the fraction of the incident infrared radiation which is absorbed by specific molecular structures which happen to absorb at that specific energy level (Stuart 2004). Such molecular structures may for example be aromatic C=C bonds, C-N bonds in amides, or C-O bonds in alcohols (e.g. carbohydrates) (Stuart 2004). When molecular structures absorb infrared radiation, this causes a change in dipole moment of one or more of their bonds and the larger the change in dipole moment, the more intense is the absorption and hence the larger the peak in a MIRS. The same stretching results in a larger change in dipole moment for more electronegative bonds than less electronegative bonds (Stuart 2004). Since C-N bonds are more electronegative than aromatic C=C bonds, this means that already a small amount of proteins in peat can contribute

equally large or more to the peaks in MIRS around 1510 and 1630 cm^{-1} than aromatic C=C bonds in the same region of MIRS (see Fig. 3 in Reuter et al. (2020) for an example). These confounding factors make it necessary to explicitly define the conditions under which a spectral prediction model or other variable derived from MIRS (e.g. a humification index) is a valid proxy for a specific, precisely defined, organic matter fraction.

One major reason why the model for Klason lignin in Hodgkins et al. (2018) is biased and invalid for peat is that the only predictor variable used in this model, `arom15arom16`, is not a good indicator for Klason lignin because, as explained above, also proteins can absorb in the same energy range (Stuart 2004) and therefore, estimates are biased depending on the amount of proteins in the samples (Teickner and Knorr 2022). Therefore, we should interpret `arom15arom16` and similar variables, such as specific humification indices ($HI_{1630/1090}$, $HI_{1510/1090}$) only then as good proxies for the relative abundance of aromatic C=C bonds if we have shown that protein contents do not differ much between the peat samples (thresholds which still need to be established).

In all other cases, where we cannot validate if a variable is a good proxy for a specific organic matter fraction, we should be precise in our wording by calling the variable `arom15arom16` (instead of Klason lignin or aromatics) to signal that this variable may be no good indicator for aromatics if it is confounded by proteins, i.e. we may agree on `arom15arom16` as precise name for this variable, but it must always be understood that it is first and foremost *only* defined as sum of the area of two peaks extracted by the specific procedure proposed in Hodgkins et al. (2018) and nothing more. Obviously, this recommendation equally applies to all other variables derived from MIRS.

Hodgkins et al. (2018) have made an important first step to actually quantify holocellulose and Klason lignin contents, i.e. specific concepts of carbohydrates and aromatics, from MIRS and this is an important improvement over the qualitative interpretation of peak heights or peak ratios (such as humification indices) used in the past (e.g. Cocozza et al. (2003), Broder

et al. (2012), Tfaily et al. (2014)). Using precise words for what we have actually measured and what confounding factors we have considered is important to assure that this first step will actually be an improvement over the qualitative interpretation of MIRS.

Conclusions

Our reanalysis of Hodgkins et al. (2018), taking into account previously unconsidered sources of uncertainties, shows that:

1. Larger aromatic contents may be necessary to stabilize tropical near-surface peat against decomposition and aromatics may accumulate at a higher rate with depth in tropical peatlands than estimated by Hodgkins et al. (2018).

2. Larger uncertainties indicate that also larger differences in aromatic contents between (sub)tropical and high latitude peat than estimated by Hodgkins et al. (2018) are possible, also between deeper peat.

3. More research should focus on how estimates of carbohydrate and aromatic contents from MIRS may be confounded by other organic matter fractions, in particular proteins. As a first step, this requires accurate concepts to name organic matter fractions and variables used in the interpretation of MIRS.

The results of this reanalysis also apply to a more recent extension of Hodgkins et al. (2018) with a larger dataset (Verbeke et al. 2022).

Supporting information

Supporting information S1 is available as appendix to this manuscript.

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Competing interests

The authors declare no competing interests.

Author contributions

HT performed the calculations, prepared the figures, and wrote the original text. Both HT and KHK revised and edited the text. KHK provided funding for this study.

Data and Code availability

Data and code to reproduce our analyses are available via https://doi.org/10.5281/zenodo.10276230 (Teickner and Knorr 2023).

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Supplementary Information S1 to: "Comment on Hodgkins et al. (2018)"

Henning Teickner[1,*], Klaus-Holger Knorr[1]

[1] ILÖK, Ecohydrology and Biogeochemistry Group, University of Münster, Heisenbergstr. 2, 48149 Münster, Germany

[*] Corresponding author: Henning Teickner (henning.teickner@uni-muenster.de)

1 Methods

For our analysis, we used three models for each holocellulose and Klason lignin:

1. The models described in Hodgkins et al. (2018) to predict holocellulose and Klason lignin contents from mid infrared spectra, but recomputed in a Bayesian framework. This is the "original Bayesian model" in Teickner and Knorr (2022). These models were used to reproduce the computations in Hodgkins et al. (2018). Here, these models are called "original" models.

2. A modified model to predict each, holocellulose and Klason lignin contents from mid infrared spectra. These are the models called "best binned spectra" models in Teickner and Knorr (2022). A brief description of these models is given in section Computation of modified models to predict organic matter holocellulose and Klason lignin contents from mid infrared spectra. These models are used to reanalyze holocellulose and Klason lignin depth profiles for the two latitude classes defined in Hodgkins et al. (2018), as

described in section Depth profiles.

3. The same models as in number 2, but extended such that it does not only predict holocellulose or Klason lingin contents, but also describes the latitudinal gradient in average near-surface holocellulose or Klason lingin contents. These models are described in detail in section Latitudinal gradients.

To differentiate the models, we call the models from number 1 "original models", the models from number 2 "modified models using only the mid infrared spectra" and "Modified, only MIRS-based" in figure 3, and the models from number 3 (whenever it is not clear from the context which model we refer to) "our models for the latitudinal gradient" and "Modified" in figure 3.

1.1 Computation of modified models to predict organic matter holocellulose and Klason lignin contents from mid infrared spectra

Computation of the modified models using only the mid infrared spectra (MIRS) is described in detail in Teickner and Knorr (2022). In brief, we used the same training data as Hodgkins et al. (2018) and a Bayesian linear regression model which we computed with Markov Chain Monte Carlo (MCMC) sampling using brms (Bürkner 2017, 2018) and rstan (Stan Development Team 2020) which are both interfaces to Stan (Stan Development Team 2021). In contrast to the original models of Hodgkins et al. (2018), we (1) used a beta distribution as likelihood function instead of assuming a Gaussian distribution, because holocellulose and Klason lignin contents cannot be negative or larger than 100 mass-%, we (2) did not use manually selected peaks extracted from the spectra, but all the z-transformed absorbance values after baseline correction, normalization, and binning (bin width = 20 cm^{-1}), and we (3) did not use simple linear regression, but regularized linear regression by using a regularized horseshoe prior (Piironen and Vehtari 2017a, 2017b) for coefficients for spectral

variables. For the intercept and for the scale parameter of the beta distribution, we used weakly informative default priors of rstanarm (Goodrich et al. 2020) and brms (Bürkner 2017, 2018).

1.2 Reanalysis of latitudinal and depth patterns, considering prediction uncertainty of the mid infrared-based models

1.2.1 Depth profiles

Predictions of the modified models using MIRS only were used to create figure 2 in the main text. For this, we extracted posterior draws from the models and for each MCMC iteration fitted a LOESS smoother with the same parameters as Hodgkins et al. (2018). From the fitted values (average predictions), we computed predictions across depth and from these 95% uncertainty intervals. We contrast these predictions and intervals with the intervals as computed by Hodgkins et al. (2018) where the prediction uncertainty of the model is not considered.

We acknowledge that this is no full Bayesian analysis. In a fully Bayesian analysis and uncertainty propagation, parameters for the smoother would have assigned prior distributions and the distribution of the parameters for the LOESS smoother would inform also the parameters of the regression model predicting holocellulose and Klason lignin contents, respectively, from the spectral data.

We did not compute a full Bayesian model here for the following reasons:

1. Since representative training data are missing, both the original and modified models cannot be fully validated and prediction uncertainties are probably larger than computed with these models (Teickner and Knorr 2022). Since representative test data are missing, these additional uncertainties cannot be estimated. We assume that these uncertainties are larger than those induced by parameter uncertainty from models for

the depth profiles.

2. The 45° latitude threshold applied in separating the data is a heuristic threshold to analyze some differences, but there is no specific reason about choosing this or some other value, except that sample numbers would be less balanced for this particular dataset. The goal therefore was not to compute a model which describes the underlying data optimally, but to show how depth profiles of the two heuristic latitude classes differ if one uses the same method as Hodgkins et al. (2018) (to compute the depth profile), but considers the prediction uncertainty of the models.

The modified models using only the MIRS were also used to compute median values and confidence intervals mentioned in the main text in section "Klason lignin content and residual enrichment in (sub)tropical peatlands probably are larger". In these computations, we also considered the predictive uncertainty of the original models (the Bayesian formulation described in Teickner and Knorr (2022)).

1.2.2 Latitudinal gradients

To reproduce the latitudinal gradients (figure 1 in the main text), we computed a hierarchical Bayesian model which is an extension to the modified model to predict holocellulose and Klason lignin contents using only the spectra, respectively. The extension is that we generate predictions for the peat samples by treating their holocellulose content (or Klason lignin content) as missing data. These predicted values are linked to a second model which is a linear regression model for the latitudinal gradient in average surface peat contents of holocellulose (or Klason lignin). Holocellulose (or Klason lignin) contents of individual peat samples are modeled with a beta distribution for each peat core. The mean value of these beta models is informed by a beta distribution describing the average surface peat content of an assumed population of peat cores. Similarly, the scale parameter of the beta models for each cores is informed by a gamma distribution describing the average scale parameter of an assumed population of peat cores. The full model for holocellulose content is:

$$
\begin{aligned}
\mathrm{Y}_1[i] &\sim \text{beta}(\mu_1[i]\phi, (1-\mu_1[i])\phi) \\
Y_2[j] &\sim \text{beta}(\mu_2[j]\phi, (1-\mu_2[j])\phi) \\
\mu_1 &= \text{logit}^{-1}\left(\alpha + \beta \mathrm{X}_1\right) \\
\mu_2 &= \text{logit}^{-1}\left(\alpha + \beta \mathrm{X}_2\right) \\
\alpha &\sim \text{normal}(0,\ 0.2) \\
\beta &\sim \text{horseshoe}(zb, hs_{local}, hs_{global}, hs^2_{scale\_slab} \cdot hs_{slab}) \\
\phi &\sim \text{gamma}(90, 1) \\
Y_2[j] &\sim \text{beta}(\mu_{\text{core}}[\text{index\_core}[j]]\phi_{\text{core}}[\text{index\_core}[j]], (1-\mu_{\text{core}}[\text{index\_core}[j]])\phi_{\text{core}}[\text{index\_core}[j]]) \\
\mu_{\text{core}}[k] &\sim \text{beta}(\mu_{\text{pop}}[k]\phi_{\text{pop}}, (1-\mu_{\text{pop}}[k])\phi_{\text{pop}}) \\
\mu_{\text{pop}}[k] &= \text{logit}^{-1}\left(\alpha_{\text{latitude}} + \beta_{\text{latitude}}\mathrm{x}_{\text{latitude}}[k]\right) \\
\phi_{\text{core}} &\sim \text{gamma}(50, 1) \\
\phi_{\text{pop}} &\sim \text{gamma}(120, 3) \\
\alpha_{\text{latitude}} &\sim \text{normal}(-0.8, 0.08) \\
\beta_{\text{latitude}} &\sim \text{normal}(0, 0.3)
\end{aligned}
$$

All variables and parameters with subscript "1" refer to the training data for the MIRS prediction model (diverse organic matter samples, including leave, wood, and paper products):

- Y_1 are the measured holocellulose contents of the training data.
- μ_1 are the respective average values modeled with MIRS.
- X_1 is a matrix containing the MIRS variables (variables in columns and samples in rows) for the training data.
- i is an index for each sample of the training data.
- ϕ is the precision parameter for the Beta distribution with which values of Y_1 are modeled.

- α and β are intercept and regression coefficients for the MIRS prediction model. The βs are modeled via a regularized horseshoe prior parameterized following Piironen and Vehtari (2017b) as described in Teickner and Knorr (2022).

All variables and parameters with subscript "2" refer to the peat samples on which the MIRS prediciton model is applied to describe the latitudinal gradient:

- Y_2 are the estimated peat holocellulose contents modeled simultaneously from the MIRS (via μ_2 — this is the same model as for μ_1) and from the average average near-surface peat holocellulose contents per core (μ_{core}).
- X_2 is a matrix containing the MIRS variables (variables in columns and samples in rows) for the peat samples.
- μ_{core} are average average near-surface peat holocellulose contents per core and is assumed to follow a Beta distribution which average value is modeled in dependency of latitude.
- μ_{pop} are average near-surface peat holocellulose contents computed for each latitude.
- j is an index for each sample in the peat data.
- index_core is a vector assigning each sample in the peat data to the respective core index.
- k is an index for each unique core in the peat data.
- α_{latitude} and β_{latitude} are intercept and regression coefficient of the linear model with which the latitudinal gradient in average near-surface peat holocellulose contents are described.
- $x_{\text{latitude}}[k]$ is the latitude for core k.
- ϕ_{pop} and ϕ_{core} are precision parameters for the gamma distributions for the average core near surface average holocellulose content and the core near surface average holocellulose contents, respectively.

The full model for Klason lignin is analogous to that for holocellulose:

$$
\begin{aligned}
\mathrm{Y}_1[i] &\sim \mathrm{beta}(\mu_1[i]\phi, (1-\mu_1[i])\phi) \\
Y_2[j] &\sim \mathrm{beta}(\mu_2[j]\phi, (1-\mu_2[j])\phi) \\
\mu_1 &= \mathrm{logit}^{-1}(\alpha + \beta \mathrm{X}) \\
\mu_2 &= \mathrm{logit}^{-1}(\alpha + \beta \mathrm{X}_1) \\
\alpha &\sim \mathrm{normal}(0,\ 0.2) \\
\beta &\sim \mathrm{horseshoe}(zb, hs_{local}, hs_{global}, hs^2_{scale\_slab} \cdot hs_{slab}) \\
\phi &\sim \mathrm{gamma}(90, 1) \\
Y_2[j] &\sim \mathrm{beta}(\mu_{\mathrm{core}}[\mathrm{index\_core}[j]]\phi_{\mathrm{core}}[\mathrm{index\_core}[j]], (1-\mu_{\mathrm{core}}[\mathrm{index\_core}[j]])\phi_{\mathrm{core}}[\mathrm{index\_core}[j]]) \\
\mu_{\mathrm{core}}[k] &\sim \mathrm{beta}(\mu_{\mathrm{pop}}[k]\phi_{\mathrm{pop}}, (1-\mu_{\mathrm{pop}}[k])\phi_{\mathrm{pop}}) \\
\mu_{\mathrm{pop}}[k] &= \mathrm{logit}^{-1}(\alpha_{\mathrm{latitude}} + \beta_{\mathrm{latitude}}\mathrm{x}_{\mathrm{latitude}}[k]) \\
\phi_{\mathrm{core}} &\sim \mathrm{gamma}(10, 1) \\
\phi_{\mathrm{pop}} &\sim \mathrm{gamma}(120, 3) \\
\alpha_{\mathrm{latitude}} &\sim \mathrm{normal}(-0.2, 0.08) \\
\beta_{\mathrm{latitude}} &\sim \mathrm{normal}(0, 0.3)
\end{aligned}
$$

Parameter values for prior distributions were chosen based on a prior predictive simulation by plotting histograms and prior model predictions within the range of the predictor values (figure 1). These plots allow to check implications of prior choices in light of the knowledge on holocellulose and Klason lignin contents available: We know e.g. from Hayes, Hayes, and Leahy (2015) that holocellulose (total sugar) and Klason lignin contents of peat in Ireland (from unknown depths) measured with a procedure very similar to that used by Hodgkins et al. (2018; De la Cruz, Osborne, and Barlaz 2016) range between 8 to 29 and 42 to 72 mass-%, respectively. Holocellulose and Klason lignin contents of peat forming vegetation (of boreal peatlands) have been reported to range between 25.8 to 70.9, and 8.1 to 54.3 mass-

%, respectively (Turetsky et al. 2008; Straková et al. 2010; Bengtsson, Rydin, and Hájek 2018). Surface peat contents should be similar to these values and holocellulose contents generally smaller and Klason lignin contents larger due to decomposition losses (Benner and Maccubbin 1984; Worrall et al. 2017). In addition, we adjusted prior choices (particularly by reducing the standard deviation of the global intercept of the model for the latitudinal gradient and other parameters (see below)) to avoid computational issues (see below). We assured via prior and posterior predictive checks that these adjustments neither result in too informative priors, nor in a bad fit to the data.

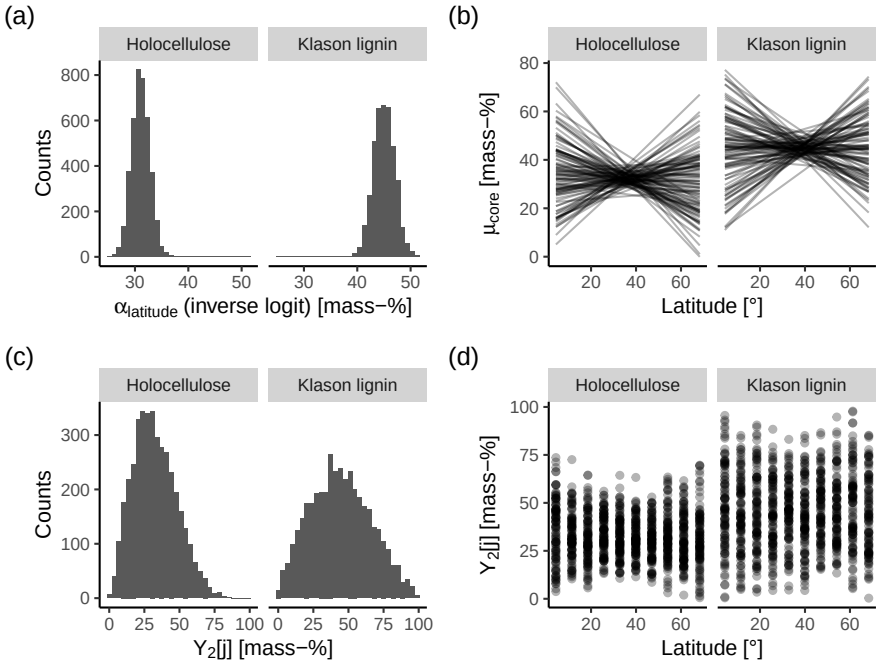

Figure 1: Prior predictive checks for the modified models describing the latitudinal gradient of peat core average near-surface holocellulose and Klason lignin contents. (a) Marginal prior distribution for the intercept of the latitudinal gradient model (average peat core average near-surface content at the average latitude of the sampled peat cores). (b) 100 random lines from the joint prior distribution for the intercept and slope for the latitudinal gradient (difference between average peat core average near-surface content at different latitudes). (c) Marginal prior distribution for the content of an individual peat sample at a latitude of ~4.4 °N. (d) At a sequence of discrete latitude values 100 randomly sampled contents for individual near-surface peat samples.

Our final prior choice assumes a population average near-surface content of holocellulose of ~30 mass-% with a standard deviation of ~2 mass-% and of Klason lignin of ~50 mass-% with

a standard deviation of ~2 mass-% (figure 1). Assuming a normal(0, 0.3) prior for β_{latitude} for both models resulted in a broad range of possible relations between contents and latitude (figure 1). Similarly, the parameter choices for ϕ, ϕ_{core}, and ϕ_{pop} resulted in reasonable ranges of contents on the level of averages for individual cores and samples, respectively (figure 1).

As mentioned above, some regularization was necessary to avoid computational issues: Less regularizing priors for α_{latitude}, ϕ_{core}, and ϕ_{pop} resulted in (more) divergent transitions and badly mixing MCMC chains. This behavior occurred because average population and core contents tended to 0 mass-%. This is a consequence of the inverse logit transformation which can cause high prior mass around 0 and 100 mass-% if the variability in its parameters is too large. We can exclude 0 mass-% as implausible and therefore can justify our regularization because it avoided a drift of the majority of the MCMC draws into implausible regions.

With this model, we could estimate the average difference in surface peat holocellulose (or Klason lignin) content for different latitudes while considering (1) the prediction uncertainty for each individual peat sample, (2) the uncertainty in the average surface peat contents for each peat core (which is heterogeneous across cores due to a finite number of samples and due to core-dependent depth gradients), and (3) parameter uncertainty of the regression model for the latitudinal gradient. In contrast, the original analysis did not consider uncertainty from predictions, and from computing average surface peat contents per core.

To create figure 1 in the main text, we used the models for the latitudinal gradient and samples from the posterior distribution to compute the predicted average and 95% confidence interval for a range of latitude values and we added predictions for the average contents for each of the sampled cores. Next, we recomputed the models from Hodgkins et al. (2018) by (1) predicting average peat sample contents with the original models (the Bayesian version described in Teickner and Knorr (2022)), (2) averaging the near surface peat sample average contents for each core, and (3) fitting an ordinary least squares regression line to these average core values. For this linear model, we plotted the confidence interval across latitude

and we added the computed average core near-surface contents. Thus, figure 1 in the main text compares the graphed values and model prediction from figure 3 in Hodgkins et al. (2018) with predictions from our modified analysis considering prediction uncertainty from the MIRS-based model part, and the averaging per core.

Additional sources for differences in predictions and prediction uncertainties in comparison to the analysis in Hodgkins et al. (2018) are the use of a beta distribution which constraints uncertainty intervals for extreme (near 0 and 100 mass-%) values, and the use of all spectral variables which resulted in reduced prediction uncertainties (Teickner and Knorr 2022).

2 Criticizing our models for the latitudinal gradient

In spite of our suggested improvements, our model is far from perfect to describe the data. One issue is that there were two divergent transitions for the model for holocellulose which probably could be avoided by using tighter priors. Another issue is that the models for the latitudinal gradient provide only little information on the variability in average surface peat contents and on the variability of individual samples. For holocellulose, it is visible that the large uncertainty in predictions and average contents of cores (near-surface) results in strong shrinkage to the populations' average (figure 4). For Klason lignin, the distribution of individual samples' contents is modeled quite accurately relative to the only-MIRS-based reference model, but there is also strong shrinkage on the level of average core contents (figure 4 (c)).

Shrinkage is generally an advantage of Bayesian hierarchical models because it improves the predictive performance of models (e.g. Park, Gelman, and Bafumi 2004; Lemoine 2019). The amount of shrinkage here is not surprising because the predicted contents of individual samples come with large uncertainties and therefore relative large shrinkage does not contradict the distributions estimated for individual samples' contents. However, in this case, shrinkage is in our opinion also an indication for underfitting. Underfitting occurs because

the latitudinal gradient describes only a small proportion of the variability in peat surface holocellulose contents. The reason is that latitude is no causal factor and that known causal factors which control vegetation and decomposition can be quite diverse on regional to local scales (figure 4). Our analysis therefore shows that we need models which explain how these known causal factors control peat holocellulose and Klason lignin contents to accurately describe their patterns on larger spatial scales.

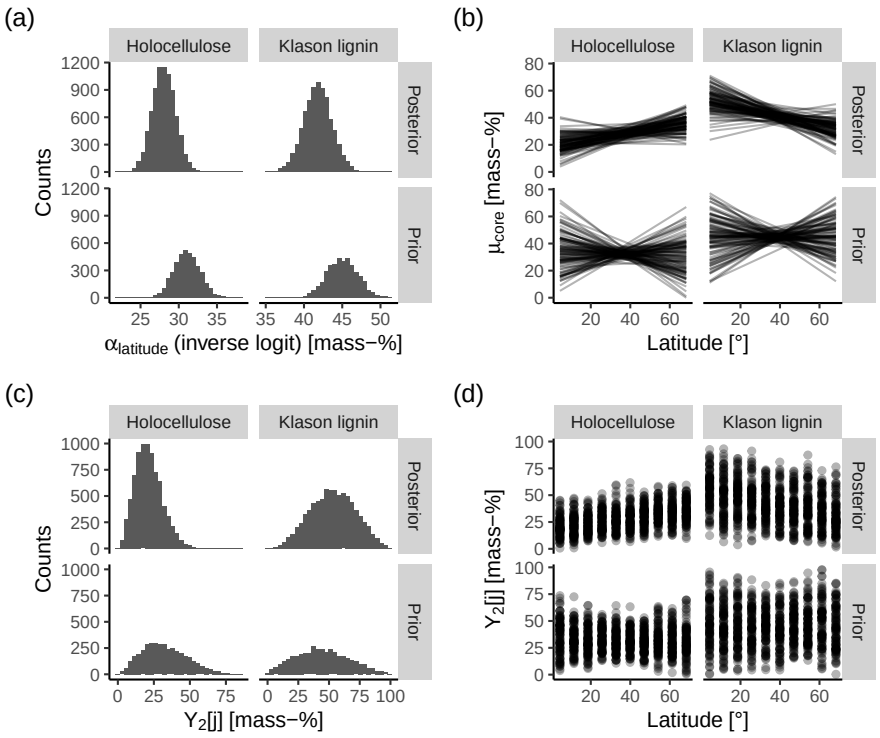

Figure 2: Posterior predictive checks for the modified models describing the latitudinal gradient of peat core average near-surface holocellulose and Klason lignin contents. The individual plots show the same as figure 1, but now additionally with the draws from the posterior distribution for the models.

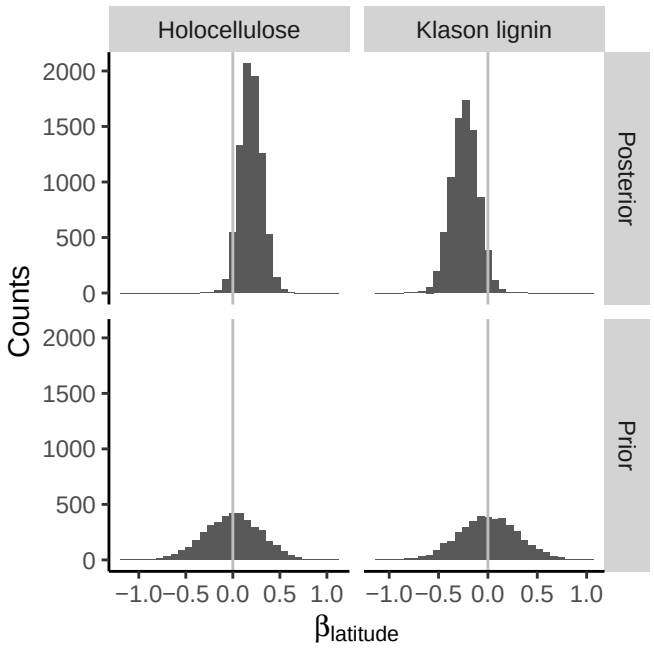

Figure 3: Marginal prior and posterior distribution for the slopes for the latitudinal gradient.

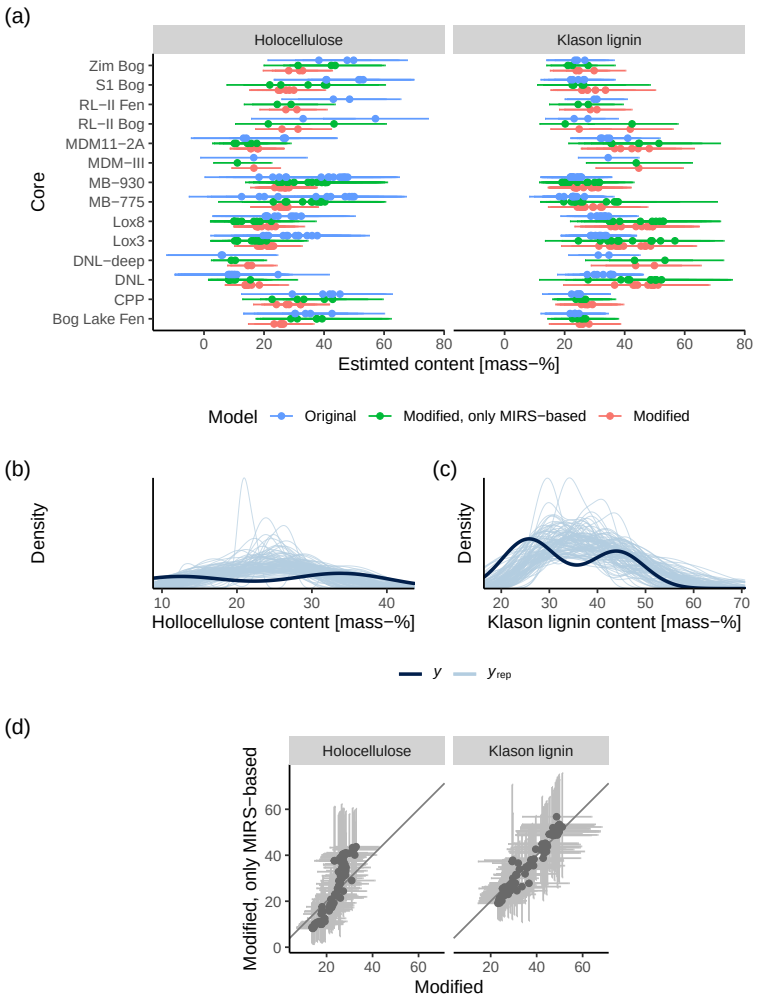

Figure 4: Shrinkage effects of the modified models describing the latitudinal gradient ("Modified") in comparison to the modified models using the MIRS only ("Modified, only MIRS-based"). (a) Predictions of the models for individual near-surface peat samples for all collected peat cores. As comparison, also the predictions of the original model (the Bayesian formulation of the models to predict holocellulose and Klason lignin contents of Hodgkins et al. (2018); "Original") are displayed. Points are average predictions and error bars 95 % prediction intervals. For holocellulose, shrinkage effects are visible both for the variability within cores and the variability across cores. (b, c) Posterior predictive densities of 100 random draws of average peat core near-surface contents (y_{rep}) in comparison to the average contents of the peat samples predicted with the modified model using only the MIRS (y). Due to shrinkage, all models produce a higher density of average peat core near-surface samples with average holocellulose and Klason lignin contents. (d) Plot of individual near-surface peat sample contents predicted by the modified models using the MIRS only versus the contents predicted by the modified models describing the latitudinal gradients. Points are average predictions and error bars 95 % prediction intervals. For holocellulose, shrinkage effects for samples with larger contents are visible.

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