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comPARAFAC: a library and tools for rapid and quantitative comparison of dissolved organic matter components resolved by Parallel Factor Analysis

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Abstract

Parallel Factor Analysis (PARAFAC) is a well-established method for characterizing dissolved organic matter (DOM). While methods for sample processing and PARAFAC analysis are well defined and robust, subsequent classification of DOM fluorescence components and comparisons of components among studies remain highly qualitative. Because these comparisons often guide the interpretation of subsequent data, it is important that quantitatively accurate comparisons be made. We developed a statistical tool, comPARAFAC, using a modified Tucker's Congruence Coefficient (mTCC), an established method of factor comparison, to provide a quantitative basis for comparing models. To develop and test this tool we used mTCC to compare factors from 35 DOM fluorescence studies using Parallel Factor Analysis (PARAFAC) in marine and freshwater environments. We compared mTCC-guided component matching with qualitative comparisons made in the literature to describe the current perceptions of component equivalence. Based on our analysis, 21% of the direct comparisons made using the qualitative approach are potentially erroneous, whereas possible matches are missed 14% of the time using that same approach. The procedure and accompanying PARAFAC model library for performing quantitative mTCC-guided comparisons are available as an R package (see Web Appendix A). This method simplifies and standardizes the process by which researchers identify and compare fluorescent DOM components across studies.

Dissolved organic matter (DOM) plays a central role in the functioning of aquatic systems. As the most abundant form of detritus, it is a key source of energy, fueling ecosystem processes in freshwater (Hall and Meyer 1998) and marine systems (Azam et al. 1983). In high abundance, DOM stimulates microbial respiration depressing dissolved oxygen levels below the level necessary to sustain aquatic life. It also filters out damaging UV radiation, which modulates phytoplankton growth in oceans (Arrigo and Brown 1996) as well as aquatic arthropod mortality in fresh waters (Williamson et al. 2001). It plays a key role in biogeochemical cycles influencing the transport and bioavailability of metals (Aiken et al. 2011; Luengen et al. 2012) and demand for nutrients (Sobczak et al. 2003). The role DOM plays in ecosystems depends on the chemical nature of the components within the DOM pool. Indeed, DOM is a highly complex pool of organic compounds with variable reactivity and bioavailability, which are frequently linked to the chemical composition of DOM (McDowell 1985; Cory and Kaplan 2012).

The sheer chemical complexity of DOM can make characterization and subsequent prediction of its ecological role difficult. Researchers have used a wide variety of methods to describe the composition of DOM. Analysis of elemental composition and element ratios are often used to infer bioavailability (Fellman et al. 2008). DOM may also be classified as "humic" or "fulvic" based on acid-base solubility. Resins may be used to separate DOM into hydrophobic (non-polar) and hydrophilic (polar) types. Specific functional groups or compounds, such as total phenolics and carbohydrates may be quantified directly (Nelson 1944; Waterman and Mole 1994).

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Infrared spectroscopy (Pernet-Coudrier et al. 2010), ^{13}C NMR spectroscopy (Mladenov et al. 2007), and high resolution mass spectroscopy (Sleighter and Hatcher 2008) have been used to define specific molecular structures and formulas present in DOM. Finally, stable and radio isotopes of C may also be used to determine source and age of DOM (Raymond and Bauer 2001; Griffith et al. 2009).

Recently, optical measures requiring minimal sample preparation have emerged as a common, cost-effective means for gaining insight into DOM composition. These methods use absorbance and fluorescence spectroscopy to characterize the chemical nature of a subset of the DOM pool. The slope of the UV-Vis absorbance spectrum can be related to bulk composition (Twardowski et al. 2004), whereas normalized absorbance at specific wavelengths (e.g., $SUVA_{254}$) have been related to molecular weight and aromaticity (Weishaar et al. 2003). Researchers have long used excitation and emission matrices (EEMs) to differentiate fluorescent DOM characteristics (Coble 1996). Excitation emission matrices are collected by reading a sample's fluorescence emission profile over a series of excitation wavelengths. This method generates a large quantity of information that must be disentangled to characterize the DOM pool. Statistical chromatography of EEMs by Parallel Factor Analysis (PARAFAC) separates overlapping fluorophores into a few key factors or "components" allowing for more detailed and quantitative usage of EEM data (Stedmon et al. 2003; Stedmon and Bro 2008).

DOM is a complex pool of molecules whose origin, age, and reactivity are difficult to determine using bulk analytical methods, such as total or dissolved organic carbon, nitrogen, and sulfur. At the watershed scale, PARAFAC allows researchers to trace the origins of DOM in rivers and estuaries (Stedmon and Markager 2005; Williams et al. 2010) and oceans (Walker et al. 2009). Much of this fluorescent pool, regardless of source, contains biologically reactive proteins and quinones that may provide C or act as terminal electron acceptors in microbial reduction oxidation processes (Cory and McKnight 2005). Within these different fractions, protein-like DOM is most frequently associated with enhanced bioavailability (Cory and Kaplan 2012) and bacterial growth (Williams et al. 2010). However, photodegradation by UV irradiation of non-labile components of the DOM pool may improve bioavailability to heterotrophs (Lu et al. 2013). PARAFAC analysis of DOM has also shown that labile DOM fractions are critical for organic N and P cycling (Lutz et al. 2012; Mineau et al. 2013) and that specific processes like denitrification may depend on organic matter quality (Barnes et al. 2012).

PARAFAC analysis is considered a robust method for analyzing composition in the fluorescent fraction of the DOM pool, and is widely used in terrestrial (Ohno and Bro 2006), marine (Stedmon et al. 2003), and freshwater studies (Holbrook et al. 2006). Once a PARAFAC model is built, researchers frequently try to assign a chemical-, source-, or process-based identity to components in the model. Many researchers have found similarities between particular components in their models and components identified in previous studies, leading them to infer sources or functional behaviors for specific components of the DOM pool (Ishii and Boyer 2012). The assignment is usually carried out by qualitative visual comparison of the shapes and excitation-emission maxima for the two components. This approach can create significant ambiguity in assigning and interpreting fluorescence characteristics and leaves readers with considerable uncertainty regarding the quantitative similarity of two components.

Such ambiguity in component matching hampers progression of the field. For example, increased use of PARAFAC inferred DOM composition is leading to meta-analysis type approaches (Ishii and Boyer 2012) without the quantitative rigor required for robust meta-analysis. For example, in an impressive review of DOM-PARAFAC literature, Ishii and Boyer (2012) identified three recurring components in 54 published PARAFAC models by qualitatively assessing similarities between line or contour plots of components. Those components were then linked to characteristics of source, physicochemical processes in aquatic systems, and treatability in engineered systems. Categorization of these components and subsequent assignments of behavior were based solely on qualitative visual comparisons. When plotted together, the actual similarity of the components in these groups appears somewhat equivocal (Fig. 1 a,b,c). Such uncertainty in these comparisons hinders broader scale syntheses of PARAFAC-derived DOM composition. A quantitative, robust, and simple method is needed for measuring the similarity in components generated across studies.

A potential solution offering a quantitative understanding of component similarity may be found using familiar distance or similarity indices. One such similarity index, Tucker's Congruence Coefficient (TCC), is used in the social sciences to compare results from factor analyses (Tucker 1951; Lorenzo-Seva and ten Berge 2006). This statistic is currently used in the DOMFluor toolbox (Stedmon and Bro 2008), a frequently used implementation of PARAFAC, to split-half validate PARAFAC models. However, it has only seen limited use in PARAFAC model comparisons in the broader literature (Murphy et al. 2008, 2011). Despite differences between studies (instrumental bias, study design, etc.) that may make model comparison difficult (Cory et al. 2010; Murphy et al. 2010), TCC can be used to quantify the degree of similarity for two factors generated in different studies over a common set of response variables or wavelengths.

Here we propose a modified TCC-based method (mTCC) and supply a library of published models for making quantitative comparisons of components among models. We used the TCC approach to gauge the strength of quantitative support for qualitative matching done in the published literature. The TCC-based method has been translated into an R package (com-PARAFAC) which provides a modified TCC as an objective, reportable, and easy to interpret statistic of similarity for the

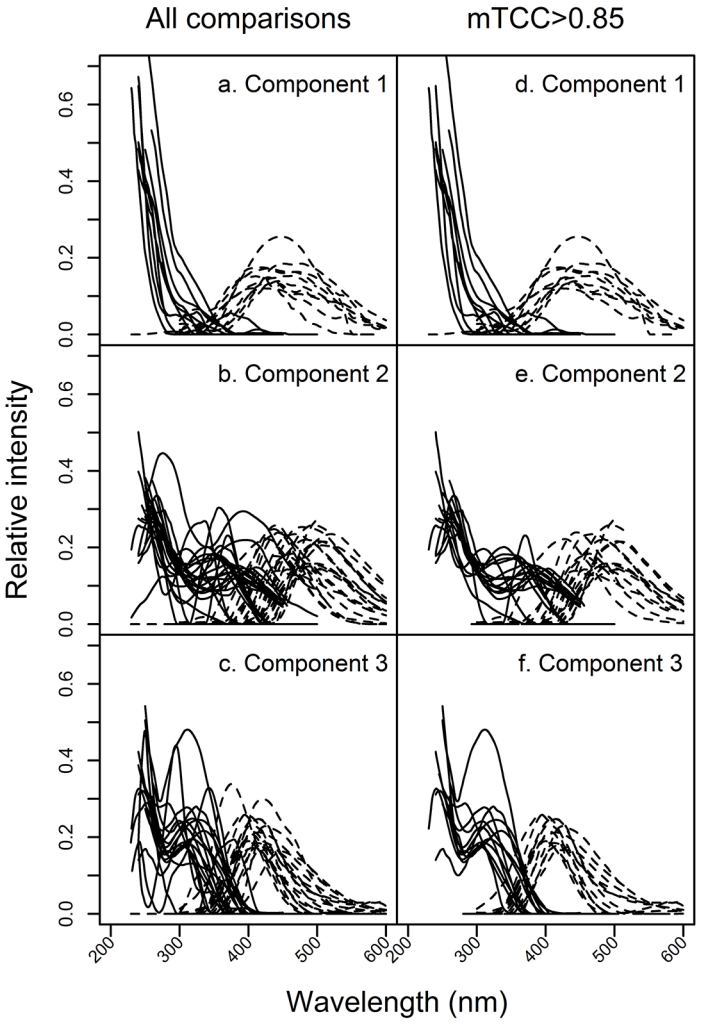

Fig. 1. Graphical representation of recurring PARAFAC components as originally grouped by Ishii and Boyer (2012) through visual assessment (panels a,b,c) and by using the average mTCC to refine component groups by removing spectra with an average mTCC < 0.85 (panels d,e,f).

quantitative comparison of components generated by different PARAFAC models. It is our hope that this will make PARAFAC analysis of fluorescence EEMs as a measure of DOM composition more accessible and interpretable for a broader array of users while reducing the uncertainty in making comparisons.

Materials and procedures

Development of this method consisted of five general steps (Fig. 2). First, published models were identified from the literature. Second, raw model data were obtained directly from the researcher or, if raw model data were not available, the models were digitized from published plots of excitation-emission spectra. Third, model wavelengths were standardized to be at equal intervals. Fourth, models were compared using Tucker's Congruence Coefficient. Finally, to evaluate subjective thresholds for equivalence, we took the subset of publications that had qualitatively compared components across models and reevaluated their comparisons based on the modified TCC.

Library selection

This comparative analysis required the development of a reference data library containing PARAFAC models from a wide range of investigations. The library was created by searching Web of Science using 'PARAFAC', 'DOM composition', and 'fluorescence' as keywords, as well as by searching for papers citing seminal works in the field (i.e., Stedmon and Markager 2005; Cory and McKnight 2005; or Stedmon and Bro 2008). Additional papers were found through communications with researchers who suggested further published data suitable for analysis. The current version of the comPARAFAC model library focuses on fresh water and marine environments, so some frequently cited models that are purely terrestrial (e.g., Ohno and Bro 2006) have been excluded. This resulted in ~ 40 candidate models. To ensure that the models included in the comPARAFAC library were as comparable as possible, model inclusion was further refined by two criteria: 1) the model must have used spectra corrected for instrumental bias (Cory et al. 2010; Murphy et al. 2010); and 2) sample preservation must not have used any compounds that alter the fluorescence spectrum (e.g., sodium azide, acidification). Omission of models from the library either reflects failure to meet these criteria or unavailability of detailed spectra for models.

Approximately 50% of the models in the library were obtained directly from researchers (Table 1); unavailable models were digitized from published figures using ImageJ (Abràmoff et al. 2004) and the Figure_Calibration plugin (Hessman 2009). Briefly, figures were calibrated to the values reported at axes ticks, then traced using a high density of points. Each excitation or emission line was reproduced using 50-100 points (~ 1 measurement every 5 nm). See Web Appendix A for detailed procedure.

Standardization of models

Quantitative comparison of models across studies requires that the models be sampled at the same wavelengths and over the same ranges. Although most studies use similar instrumental settings (excitation ~ 250- ~ 450 nm and emission ~ 250- ~ 600 nm), they may sample at slightly different wavelengths (e.g., 450 versus 452 nm). While the difference in fluorescence between these two values is small, the available quantitative tools are not valid when comparing different wavelengths. Consequently, we adjusted models to standardized increments.

Sampling wavelengths were standardized by fitting measured data with a locally weighted polynomial regression (locally weighted scatterplot smoothing [LOESS], Cleveland et al. 1992) in R (R Core Team 2012). In LOESS, at each point in the dataset, a subset of data are fitted with a low degree polynomial using weighted least squares (WLS). WLS gives more weight to points closer to the point being estimated (Cleveland et al. 1992). The size of the subset used determines the

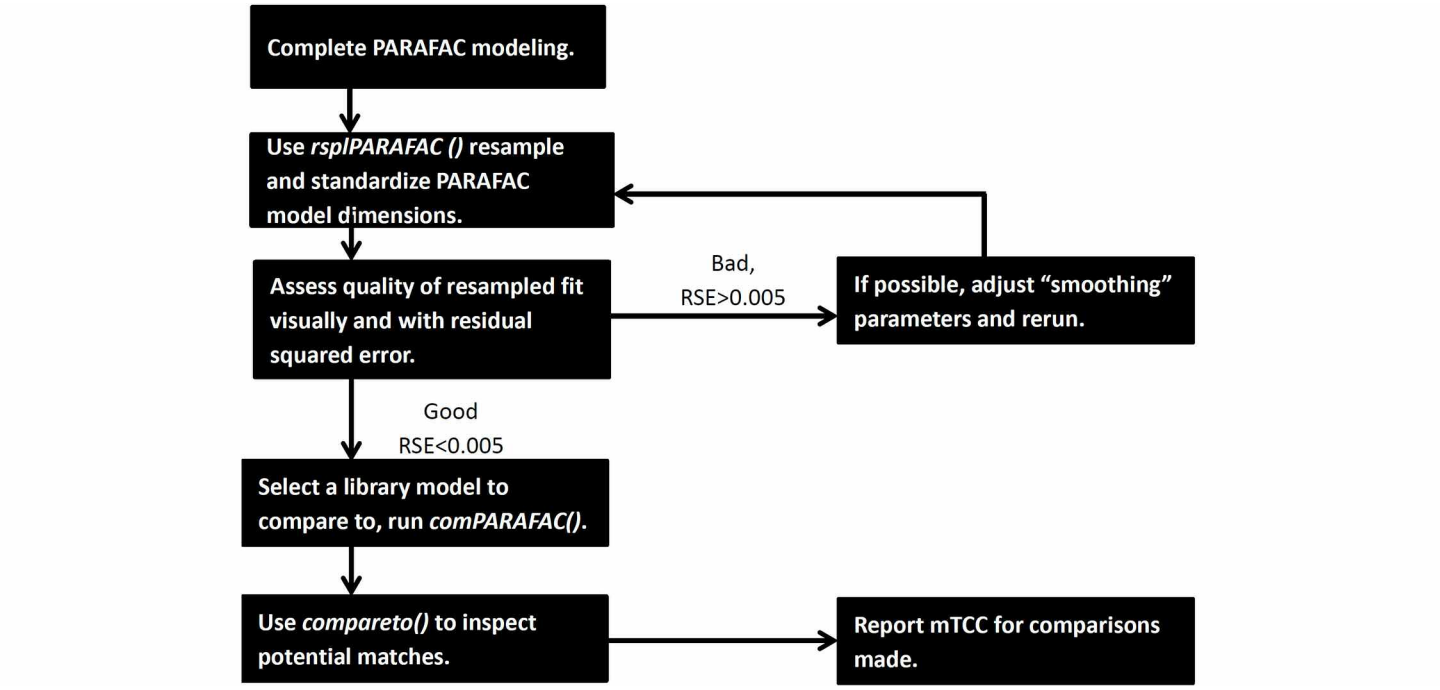

Fig. 2. Flow diagram of model comparison procedure proposed in this paper.

Table 1. Table of 37 PARAFAC model accessible in comPARAFAC library indicating their library names as well as whether they were derived from original data or transcribed from publications. For the source column, "I" indicates that it was collected with ImageJ and "O" indicates it is the original data.

Object name	Source	Citation	Object name	Source	Citation
Burrows_2013	O	(Burrows et al. 2013)	Osburn_2012	O	(Osburn et al. 2012)
Chen_2010	I	(Chen et al. 2010)	Singh_2010	I	(Singh et al. 2010)
CM_2005	O	(Cory and McKnight 2005)	Singh_2013	O	(Singh et al. 2013)
Cory_2012	O	(Cory and Kaplan 2012)	Stedmon_2003	O	(Stedmon et al. 2003)
Fellman_2009	I	(Fellman et al. 2009)	Stedmon_2005	O	(Stedmon and Markager 2005)
Fellman_2011	I	(Fellman et al. 2011)	Stedmon_2007_EST	O	(Stedmon et al. 2007b)
Guo_2011	I	(Guo et al. 2011)	Stedmon_2007_MC	O	(Stedmon et al. 2007a)
Holbrook_2006	I	(Holbrook et al. 2006)	Stedmon_2011_JGR	O	(Stedmon et al. 2011)
Hong_2011	O	(Hong et al. 2011)	Walker_2009	I	(Walker et al. 2009)
Jorgensen_2011	I	(Jørgensen et al. 2011)	Williams_2010	I	(Williams et al. 2010)
Kothawala_2012	O	(Kothawala et al. 2012)	Yamashita_2010	I	(Yamashita et al. 2010b)
Kowalczuk_2010	I	(Kowalczuk et al. 2010)	Yamashita_2010_DSRII	I	(Yamashita et al. 2010a)
Lapierre_2009	I	(Lapierre and Frenette 2009)	Yamashita_2011	I	(Yamashita et al. 2011a)
Lu_2013	O	(Lu et al. 2013)	Yamashita_2011_Odyn	I	(Yamashita et al. 2011b)
Lutz_2012	I	(Lutz et al. 2012)	Yang_2011	O	(Yang et al. 2011)
Massicotte_2011	I	(Massicotte and Frenette 2011).	Yang_2012_MC	O	(Yang et al. 2012)
Murphy_2006	O	(Murphy et al. 2006)	Yang_2013_AG	O	(Yang et al. 2013a)
Murphy_2008	O	(Murphy et al. 2008)	Yang_2013_MC	O	(Yang et al. 2013b)
Osburn_2011	O	(Osburn and Stedmon 2011)			

extent to which the regression smooths the data. A small subset will result in something closer to a point-to-point regression whereas a large subset will remove much of the variation in the original data. For our data, a typical subset was 15% of the total dataset.

The fit of the adjusted model to the original model was evaluated in two ways. First, fit was visually evaluated by overplotting original data and the fit model. Where the adjusted

model fit was visually poor, a smaller or larger subset was used. The minimum subset was 7% and the maximum subset was 25%. A smaller subset better fits sharp or irregular features, whereas a larger subset may deal better with abrupt changes in curve slope. After visual inspection, the residual squared error of the fit model was checked to ensure it was less than 5% of the maximum value of data. In practice, residual squared error (RSE) was generally less than 0.005 for emission lines, but sometimes higher (~0.01) for excitation loadings despite a good visual fit. After evaluating model fit, we resampled the models at a 1 nm interval using a LOESS regression to predict the values over the original range of data. This generated the standardized PARAFAC models in the package library.

We sought to modify the original data presented in the library as little as possible so all PARAFAC models in the library contain their original data range (approximate ± 5 nm for digitized models). Data ranges are standardized on a comparison-by-comparison basis within comPARAFAC. If the ranges scanned in two studies are different, this may result in a loss of ~50 nm at either end of the spectrum for the study scanning a wider range.

Model comparison

Once models were standardized, we used a modified Tucker's Congruence Coefficient (mTCC) to compare excitation and emission loadings (factors) from different PARAFAC models. Tucker's Congruence Coefficient is originally defined as:

$$\mathrm{TCC} = \frac{\sum x_i y_i}{\sqrt{\sum x_i^2 \sum y_i^2}} \quad (1)$$

where x is the first loading, y is the second loading, and i denotes the specific wavelength being compared. Unfortunately, this approach produces a TCC for the excitation ($\mathrm{TCC_{ex}}$) and a TCC for the emission ($\mathrm{TCC_{em}}$), which, in many cases, are disparate. Thus to determine a match and to provide a reporting statistic, we report the mTCC defined as:

$$\mathrm{mTCC} = \sqrt{\mathrm{TCC_{ex}} \times \mathrm{TCC_{em}}} \quad (2)$$

Interpretation of the TCC has been criticized as subjective. Tucker originally suggested that values from 0.98-1.00 are 'nearly exact' matches, 0.92-0.98 are a 'good' matches, 0.82-0.92 are 'borderline' matches, 0.68-0.82 are 'poor' matches, and below 0.68 are 'terrible' matches (Tucker 1951). In a critical test of the methodology, Lorenzo-Seva and ten Berge (2006) compared the visual determination of congruence by factor analysis practitioners with TCC values and concluded that > 0.95 is nearly an exact match whereas anything from 0.85-0.95 is a fair match. Within the DOMFluor v. 1.7 toolbox commonly used to implement PARAFAC analysis, the split-half validation process validates split halves by considering factors with a TCC greater than 0.95 a match.

Criteria for model matches

Similar to Lorenzo-Seva and ten Berge (2006), we evaluated PARAFAC practitioners' qualitative approach to component matching against the TCC approach to matching. We accomplished this by relating published qualitative comparisons to quantitative comparisons using mTCC. If a comparison was made to a model, it was assumed that the researcher was aware of the model, had reviewed all components in the model, and reported only the components deemed matches.

Between a model with n components and a model with m components, there are $n \times m$ possible comparisons including both matches and non-matches. For our study, if an author compared a component from their model to one in another model, we assumed this meant it meets their qualitative criteria for a match and recorded it as a "1." If no explicit comparisons were made between the other possible comparisons, these were encoded as 0. For example, if a researcher with a 5 component model compared it to a previously published 5 component model and found that 3 components matched, we would have 22 "0" and 3 "1" values. If the researcher used vague language (e.g., "this component has been found in other studies…"), we interpreted this to mean that the researcher was not confident in the comparison and did not consider it a good match; therefore, we encoded this as 0. We note that a given researcher may have considered some models that yielded no matches; if such a non-matching model was not reported by the author, we did not assume that it was part of the original comparison by the author. Thus, models without at least one component cross-matched were excluded from our analysis.

R package

All data and PARAFAC libraries used in this study, including metadata, are available for free in the R package comPARAFAC2. The package contains three functions: *comPARAFAC()*, *rsplPARAFAC()*, and *compareto()*. *comPARAFAC* compares two standardized models using a set data format that can be generated for any model using *rsplPARAFAC* (*see* Web Appendix A for details). *rsplPARAFAC* is a user-friendly model standardization algorithm that creates the standardized object used by *comPARAFAC*. Users wishing to compare a new PARAFAC model to a library model will need to first use *rsplPARAFAC* to standardize their PARAFAC model. (*See* Web Appendix A for detailed procedure and scripts). *Compareto* is a simple graphing tool allowing the user to compare two specific components. A similar function is available in *comPARAFAC*.

Assessment

Fidelity of LOESS and digitization

Data from Stedmon and Markager (SM, 2005) were used to assess the loss of fidelity between the researcher-supplied data (hereafter "original data") and data collected with ImageJ data as well as between LOESS smoothing and original data. Qualitative comparison of original data and ImageJ traces for SM1 and SM3 (Fig. 3) shows that the trace in ImageJ reproduces the original data nearly exactly. There was a minor loss of fidelity at sharp changes in the slope of the data (e.g., SM8 in Fig. 3). This may be due to low resolution or pixilation of the traced

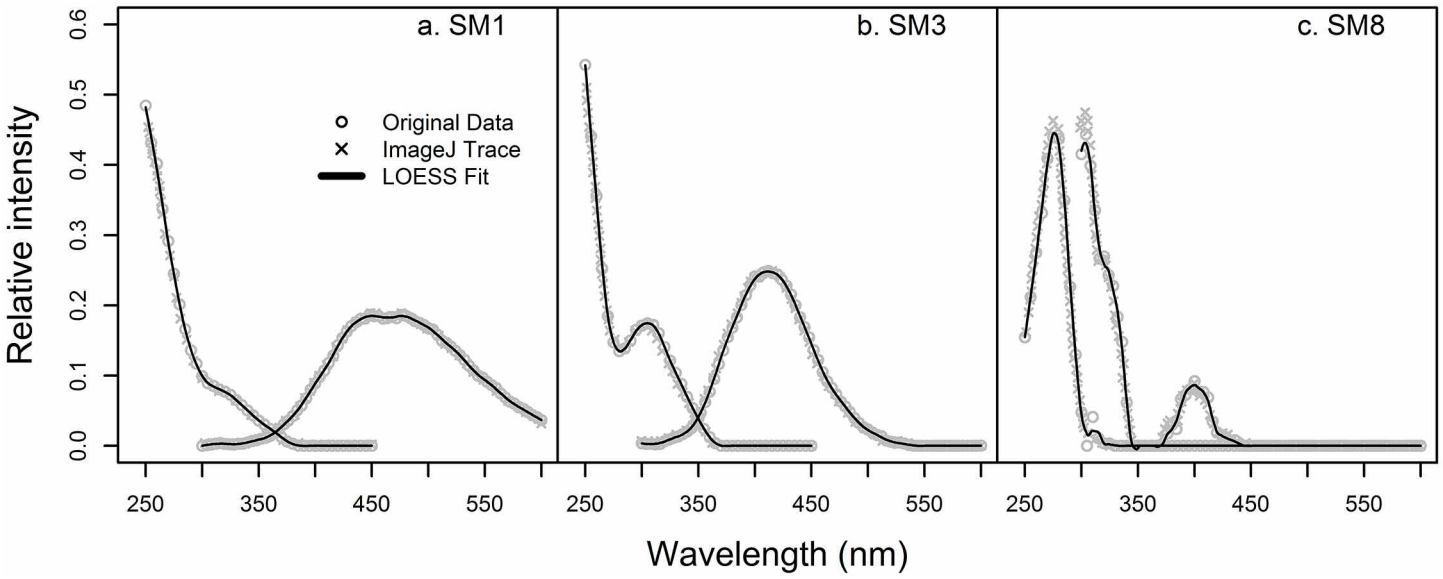

Fig. 3. Illustration of data fidelity between (a) original data, (b) ImageJ trace, and (c) LOESS regression using Stedmon and Markager (2005). In (a) and (b), the traces from the published figure and the original data are nearly identical. In SM8, there is minor loss of fidelity where the slope of the data changes sharply.

figure. The minor differences did not change the comparison between the trace and original data (Fig. 3c, mTCC = 1).

Fidelity between LOESS and the original data can be assessed using the residual standard error and by comparing both sets of data to other components. For SM1, SM3, and SM8, the residual standard error was generally less than 0.005 (ex/em: SM1 0.003/0.001; SM3 0.002/0.001; SM8 0.007/0.006, Fig. 3a). Similarly, digitized and original data returned the same mTCC value when compared with other components (e.g., comparison to component 6 in Osburn and Stedmon [2011] resulted in 0.93 for both).

Effects of instrument sampling and LOESS prediction interval on mTCC

We assessed the effect of the 1 nm resampling interval in comPARAFAC on the mTCC value by calculating mTCC for the same model sampled at 1, 2, 5, and 10 nm excitation and emission intervals. On average, the mTCC increased slightly with increasing sampling interval (~0.0003 units nm^{-1}, $P < 0.05$). However, the maximum increase from a 1 to 10 nm interval only increased mTCC by 0.3% (e.g., an mTCC of 0.9000 versus 0.9027). It is unlikely that such a change would alter the conclusions drawn from mTCC.

Although we did not assess it, a wide sampling interval may make two components (or spectra) appear more similar if the interval is large enough to skip regions of variation. Because instrumental sampling intervals are typically 5-10 nm for excitation and 2-5 nm for emission, similarity or dissimilarity may be more valid for emission than excitation spectra with large (10 nm) sampling intervals.

Qualitative comparisons versus quantitative comparisons

We used existing literature comparisons to assess the qualitative approach to component matching and to see if clear thresholds exist for matches. Our analysis revealed that the qualitative approach often is consistent with a reasonably good quantitative match (i.e., Fig. 4, a TCC > 0.90). Fig. 5a indicates that investigators mostly assign matches at an mTCC of 0.80 or greater, although some matches are assigned at much lower mTCC values (Fig. 5a). Interestingly, researchers also dismiss many matches that are above a similar mTCC threshold (Fig. 5b, 22% > mTCC of 0.80).

If the null hypothesis is that two peaks are not a match (mTCC < 0.85) and the alternative is that they are a match (mTCC ≥ 0.85), then a Type I error is falsely assigning a match (false positive), whereas a Type II error is failure to detect a match when a match is present (false negative). In our dataset, when a qualitative comparison was made (Fig. 5a), the potential rate of false positives was 21%. This occurred across 30% of the papers in which matches were assigned. Conversely, the rate of missing a quantitatively strong match (i.e., mTCC > 0.85) was 14% (Fig. 5b) and occurred across 49% of the papers surveyed. Such error rates considerably exceed the 5% threshold usually applied in statistical tests.

The occurrence of false positives and false negatives may be due to a lack of digital PARAFAC model data for making direct comparisons and the sheer number of comparisons that may be made (see Fig. 3 and Table 2 as examples). For a false negative, even with digital model data, a researcher is likely to report only the 'best' match (e.g., if selecting between an mTCC of 0.93 and an mTCC of 0.97, the researcher may only report the 0.97). Missed matches are attributable to researcher discretions. They may feel that a match does not reflect the goals of the study, which represents lost opportunities for

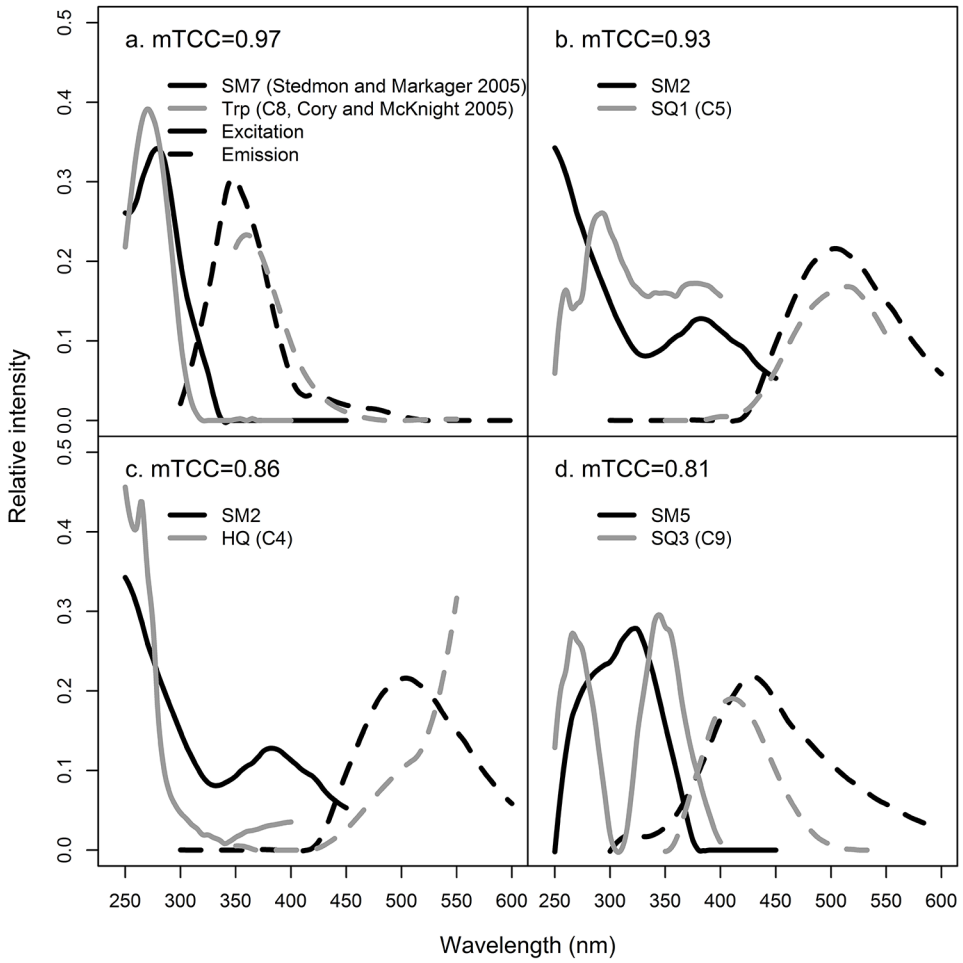

Fig. 4. Hypothetical comparisons of components observed in Stedmon and Markager (2005) with peaks found in Cory and McKnight (2005) at 4 levels of mTCC. For clarity and consistency, we refer to components from the Stedmon and Markager model with the prefix SM and components from the Cory and McKnight with the names assigned in that paper: their PARAFAC model component numbers are prefixed with C and referred to by the chemical structures tryptophan-like (TRP), semi-quinone (SQ), and hydroquinone (HQ) assigned by Cory and McKnight 2005. (a) Tryptophan-like SM7-TRP/C8, (b) SM2 – SQ1, (c) SM2 – HQ, (d) SM5-SQ3. Note: the calculation of the mTCC coefficient is only based on the excitation or emission regions that overlap.

insight; or, they may feel that two components, which are quantitatively similar, are not qualitatively similar due to small but critical spectral differences. The above calculation of type II error (14%) does not distinguish between these two situations and may be inflated.

False matches (type I errors) are more serious when comparing between PARAFAC components. When a false match is made, there is a potential for erroneous conclusions to be drawn about the composition or behavior of a component. Such errors can propagate into subsequent literature potentially affecting interpretations of components in future studies.

The utility of a quantitative approach is demonstrated by comparing components in models constructed by Stedmon and Markager (SM, 2005) and Cory and McKnight (CM, 2005) (Fig. 4). SM and CM are two models from different systems and instruments, yet they resolve what appear to be similar components. For example, both studies report tryptophan-like components (SM2 and C8/TRP, following naming conventions in the original paper), which are a strong match qualitatively and quantitatively (mTCC > 0.95, Fig. 4a). Fig. 5b illustrates another comparison between SM2 and SQ1. These two components are similar in emission and longer excitation wavelengths, but clear spectral differences are present at shorter excitation wavelengths resulting in a somewhat lower mTCC of 0.93. SM2 was described as a terrestrial or autochthonous fulvic acid-like fluorophore present in all environments (Stedmon and Markager 2005), while SQ1 was described as a semi-quinone (SQ) with terrestrial plant origins correlated to other (nonfluorescent) indicators of terrestrial organic matter (Cory and McKnight 2005). Given the similarity between peaks, the general agreement by both authors of a potential terrestrial origin for this peak, and the calculated mTCC value of 0.93, this represents a second example of a match that is

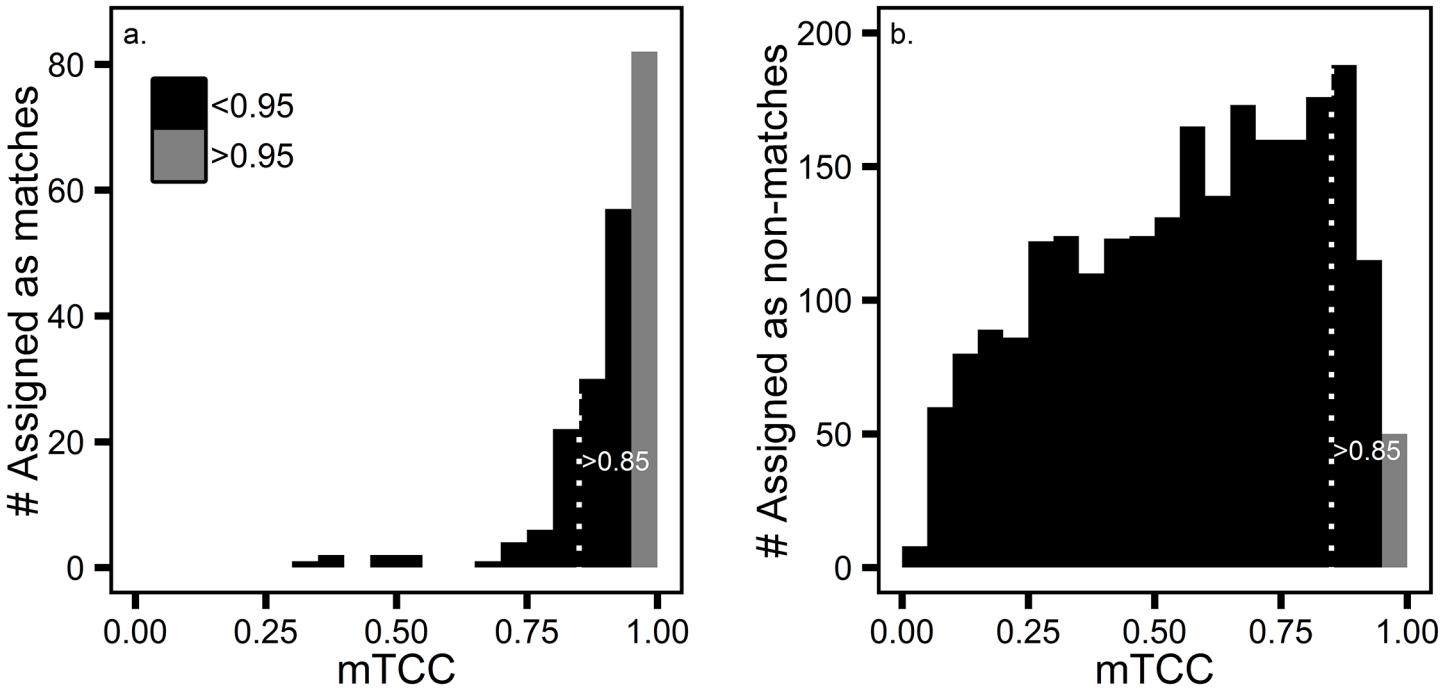

Fig. 5. Histograms summarizing results of the literature survey for qualitative comparisons that were explicitly made (a, $n = 203$) and qualitative comparisons that were implicitly not made (b, $n = 2379$) by researchers versus calculated mTCC values. Bin widths are 0.05 in both cases. In (a), researchers typically make qualitative comparisons at an mTCC greater than 0.95 only 39% of the time. In (b), note that researchers overlooked ~ 50 potentially strong comparisons.

Table 2. mTCC ranges and values within reoccurring PARAFAC component groups identified by Ishii and Boyer (2012). The mTCC range encompasses comparisons among all components assigned to a specific group (see Table S1 in Ishii and Boyer 2012). The proportion of comparisons in a given range is the frequency that two components in a group had an mTCC in that range. The average mTCC ($\overline{\text{mTCC}}$) within a group was refined by removing individual components with low pairwise $\overline{\text{mTCC}}$.

Proportion of comparisons made within given mTCC range									
Reoccurring group	**mTCC range**	**<0.80**	**0.80–0.85**	**0.85–0.90**	**0.90–0.95**	**0.95–1.00**	**nr compared**	**$\overline{\text{mTCC}}$ initial**	**$\overline{\text{mTCC}}$ refined**
Component 1	0.64-0.99	0.11	0.11	0.14	0.30	0.34	44	0.93	0.95
Component 2	0.55-0.99	0.20	0.11	0.17	0.24	0.28	264	0.88	0.94
Component 3	0.44-0.99	0.25	0.16	0.16	0.23	0.20	299	0.85	0.91

supported by both qualitative visual and quantitative statistical evidence.

We also tested the utility of this approach in improving discrimination among potentially matching components in meta-analyses using 20 of 54 models reviewed and categorized by Ishii and Boyer (2012). To do this, we calculated the mTCC matrix among the components in each qualitatively defined group. We then removed components with an average mTCC < 0.85 and replotted the data. Visually, component group 1 (Fig. 1a) represents a clear group and reanalysis of these components using mTCC only resulted in removal of one component (Fig. 1d). Components 2 and 3 (Fig. 1b, c) qualitatively present a less well-defined group. This is quantitatively confirmed (Table 2) with mTCC values for pairwise within-group comparisons greater than 0.90 for only 52% and 43% of the matches for components 2 and 3, respectively. Furthermore, 20% to 25% of the original within group comparisons had an mTCC less than 0.80. Filtering matches by average mTCC resulted in the removal of nine components peaks from component group 2 and six components from component group 3. For component group 2, this process improved the average group mTCC from 0.88 to 0.94, and in group 3, the average mTCC improved from 0.85 to 0.91 (Table 2). The resulting groups are now visually clearer (Fig. 1d, e) and quantitatively supported.

Discussion

comPARAFAC provides a simple, standardized method to quantify the uncertainty in component matching across published PARAFAC models. It also reduces the time that researchers need to spend looking for possible matches by first

guiding researchers toward the best statistical comparisons. This enables researchers to invest more time in interpreting what the similarities mean as opposed to whether two components are truly similar.

Quantitative comparisons are a key requirement for productive meta-analyses of PARAFAC data. Despite the large number of published PARAFAC models, meta-analyses have thus far remained qualitative and reliant on subjective human determination of "similarity." Our analysis of qualitative matching suggests that whereas such matches are frequently strong, failures to distinguish strong matches and inclusion of poor matches occur with reasonable frequency. Quantitative tools disambiguate model comparison and permit quantification of certainty in component matching. The ability to perform meta-analyses on published DOM composition studies may open new insights into global carbon biogeochemistry and anthropogenic influences.

The mTCC approach is not intended to replace visual inspection, and it does not yield definitive assignment of component matches; rather it is a tool that helps quantify similarity between components. We suggest an mTCC threshold or individual values should be reported when practitioners make direct comparisons to previous models to demonstrate quantitative strength of match. We found a wide range in mTCC values (0.70-0.99) over which researchers historically have assigned and dismissed matches. In this article, we suggest a more constrained set of thresholds (0.85, 0.90, and 0.95) to gauge strength of a "match." Based on what the literature has previously defined as a match when evaluating qualitative comparisons (Fig. 5), if proposed qualitative comparisons have an mTCC > 0.95, they should be considered strong matches while components with an mTCC between 0.90 and 0.95 may be considered good matches. Candidate matches with values of 0.85-0.90 are questionable. Such categories, of course, are qualitatively defined, but mTCC provides a quantitative indication of the strength of match that, combined with visual inspection, strengthens assertions of similarity between components. We expect that as mTCC is more broadly used, thresholds for strength of match may be more clearly assigned, and this may be complemented by other criteria as new tools emerge.

Existing guidance (Tucker 1951; Lorenzo-Seva and ten Berge 2006), primarily from the social sciences, may not be strictly appropriate for interpreting fluorescence components. When looking across datasets, variables such as instrument used (Cory et al. 2010; Murphy et al. 2010), pH, or salinity may require that we relax the TCC values considered for a match. At the same time, the need to relax TCC values to obtain matches in model comparisons, may be a good indicator that the models should not be compared.

Comments and recommendations

It is crucial that researchers using this package view it as a tool to help guide comparisons and improve certainty in reporting matches. Whereas the mTCC may provide an excellent indicator of a statistical match, it does not necessarily provide insight as to whether the study design or environmental conditions underlying a published model are appropriate for the comparison. Use of this package requires installation and a minimal knowledge of R (we recommend that you also install "R Studio" interface for R). All procedures and code for using this package are explained in a detailed tutorial found in Web Appendix A. It is our intention to update this package on an annual basis or when sufficient models have been received. Our analysis has used data from aquatic habitats, but the mTCC approach and data within comPARAFAC can be used across terrestrial and aquatic systems. We encourage use of comPARAFAC in a broad array of ecosystems and hope to include data from such systems in future versions of the library. Because we likely overlooked some published models, we also encourage researchers whose models fit the criteria, but were overlooked, to submit their models. Future releases will be made available through the Comprehensive R Archive Network (CRAN) at http://cran.r-project.org/.

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