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PARAFAC2: A Core Perspective

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Overview

- Introduction and motivation for research
- Relevant prior work
- PARAFAC2 and Core-PARAFAC2 Algorithms
- Imposing Nonnegativity
- Analysis on Simulated Data
- Analysis on Fluorescence Data
- Results
- Conclusions and future directions

Introduction

- Numerous R&D areas employ fluorescence lifetime to characterize fundamental molecular properties.
- Multiway fluorescence measurements provide greater analytical power than single modalities
- Two-way wavelength-time matrices (WTM) collected at different excitation wavelengths can produce three-way data
  - Time-Resolved Excitation Emission Matrix (TREEM).
- PARAFAC is an excellent data analytical tool for fluorescence data, but is limited when the data do not meet the strict expectations of trilinearity
- PARAFAC2 can accommodate shifts or scale differences in one modality and overcome restrictions
- Core PARAFAC2 allows easy implementation of nonnegativity constraints in all three modes

Motivation

- PARAFAC2 is an important multiway analysis tool when PARAFAC is inappropriate.
  - One mode has axes that are not aligned or inconsistent
  - Multiple hyphenated chromatographic profiles are a prime example
- PARAFAC2 can be slow, and difficult to implement with large data sets
- Nonnegativity constraints are difficult to apply in all three data modes
  - "…, it is impossible to impose non-negativity constraints on the [varying mode],…"*
- Improving PARAFAC2 may facilitate its broader uses in data analyses

*J. M. Amigo, et al., "Solving GC-MS problems with PARAFAC2," TrAC, Trends Anal. Chem. 27(8), 714-725 (2008)

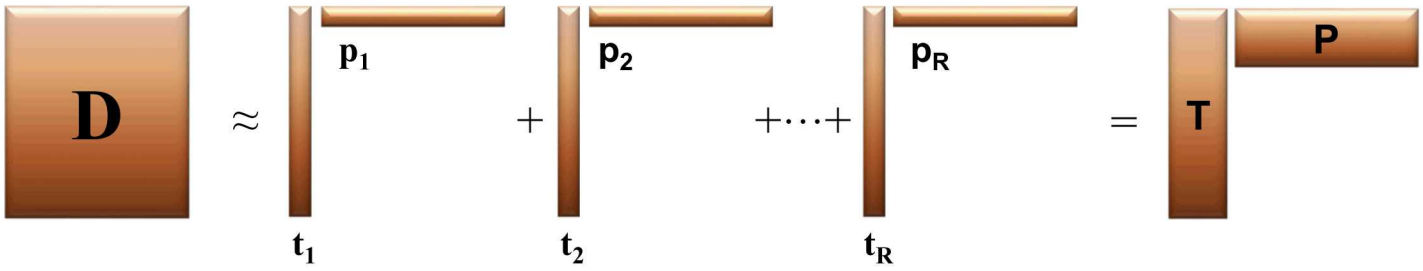
Relavent Published Research

- H. A. L. Kiers, J. M. F. TenBerge and R. Bro, "PARAFAC2 - Part I. A direct fitting algorithm for the PARAFAC2 model," J. Chemom. 13(3-4), 275-294 (1999)
- R. Bro, C. A. Andersson and H. A. L. Kiers, "PARAFAC2 - Part II. Modeling chromatographic data with retention time shifts," J. Chemom. 13(3-4), 295-309 (1999)
- J. M. Amigo, et al., "Solving GC-MS problems with PARAFAC2," TrAC, Trends Anal. Chem. 27(8), 714-725 (2008)
- M. H. Van Benthem and M. R. Keenan, "Tucker1 model algorithms for fast solutions to large PARAFAC problems," J. Chemom. 22(5), 345-354 (2008)

Recent Publications Describing PARAFAC2 with Constraints

- A. Afshar, et al., "COPA: Constrained PARAFAC2 for Sparse & Large Datasets," in eprint arXiv:1803.04572, ARXIV (2018).
- J. E. Cohen and R. Bro, "Nonnegative PARAFAC2: a flexible coupling approach," in eprint arXiv:1802.05035, ARXIV (2018).

Two-Way Analysis Methods

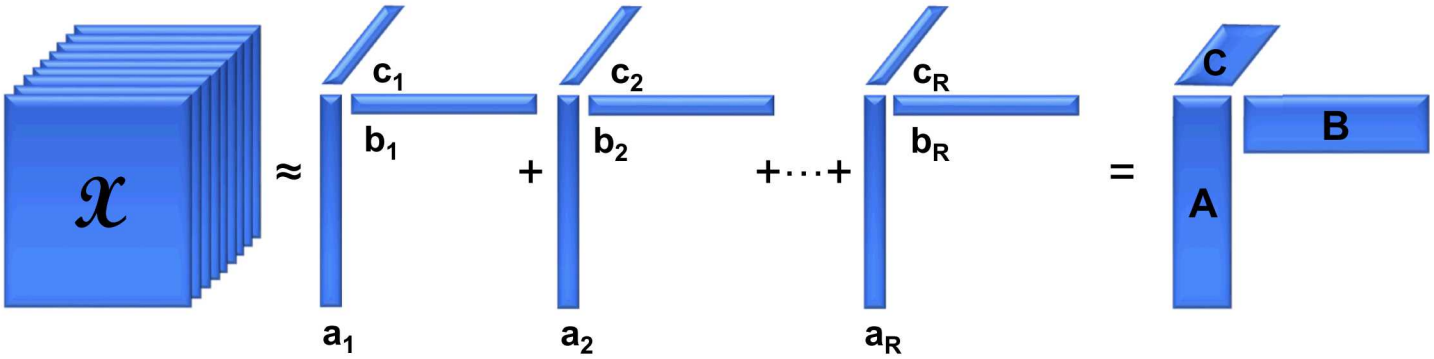


- Principal Component Analysis (PCA)
  - Given a matrix containing data, D, as a first step in many analyses we want principal components

    $$\mathbf{D} \cong \mathbf{TP}^{\mathbf{T}}$$

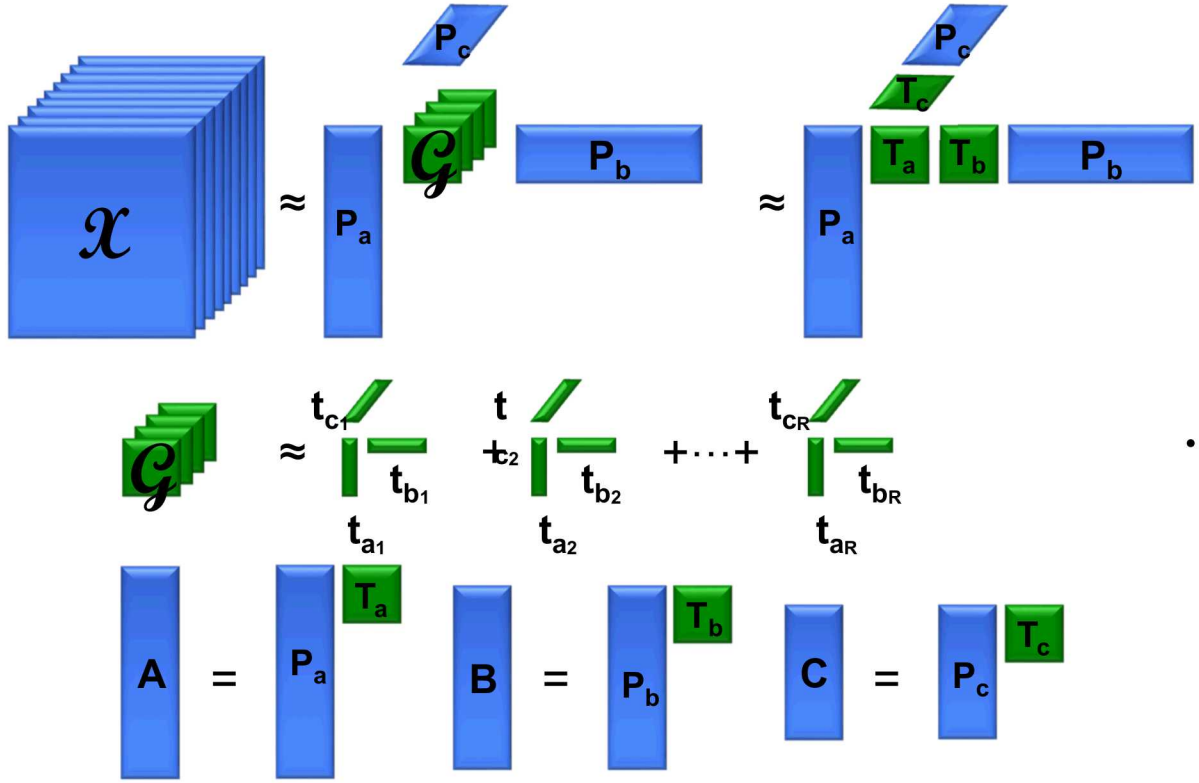
  - Such that T and P are an orthogonal basis sets, that is a reduced dimensional representation of D, with ordered maximized variance.
    - T is orthogonal (scores); P is orthonormal (loadings).
- Multivariate Curve Resolution (MCR)
  - Impose constraints on solution space

Tensor Factorization-PARAFAC



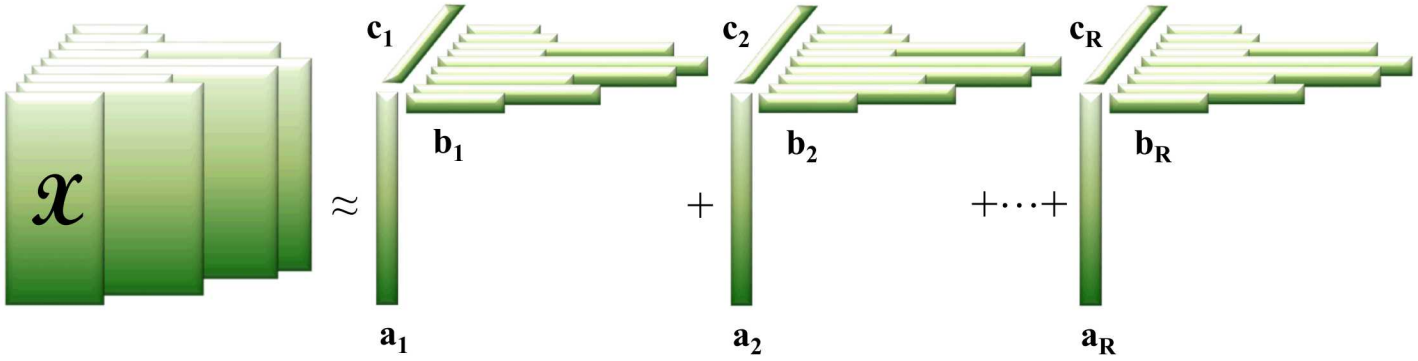
- Tensor factorizations of multi-way data
  - Parallel factor analysis (PARAFAC)
  - Nonnegative tensor factorization (NTF)
  - Similar in idea to least squares matrix techniques: principal component analysis (PCA), singular value decomposition (SVD), multivariate curve resolution (MCR)
- When applied to a data array, data are modeled as a mixture of factors, each with its own triad of signature factors

Tensor Factorization-Core PARAFAC



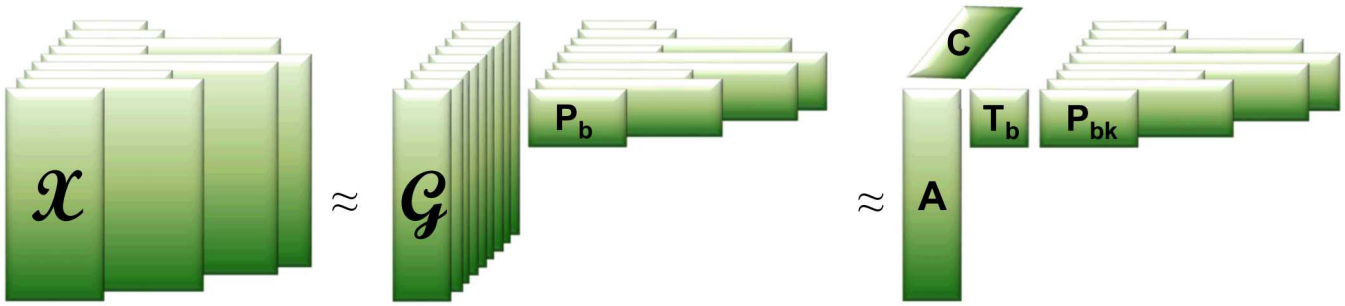
- Tensor factorizations of multi-way data
  - Tensor is initially factored using a 3-way orthogonal decomposition to generate orthogonal factors and core
  - Parallel factor analysis (PARAFAC) is performed on core to generate rotation-matrix factors

Tensor Factorization-PARAFAC2

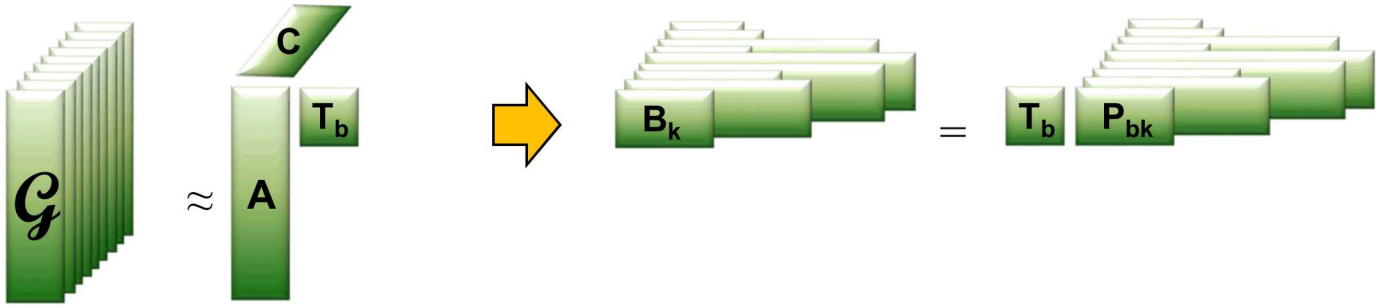


- PARAFAC2 permits factorization of unaligned or different length signals
- This data appears to not follow a model suitable for PARAFAC
  - Temporal alignment of sinus rhythms not trivial
  - Lack of alignment may prevent use of standard PARAFAC algorithm

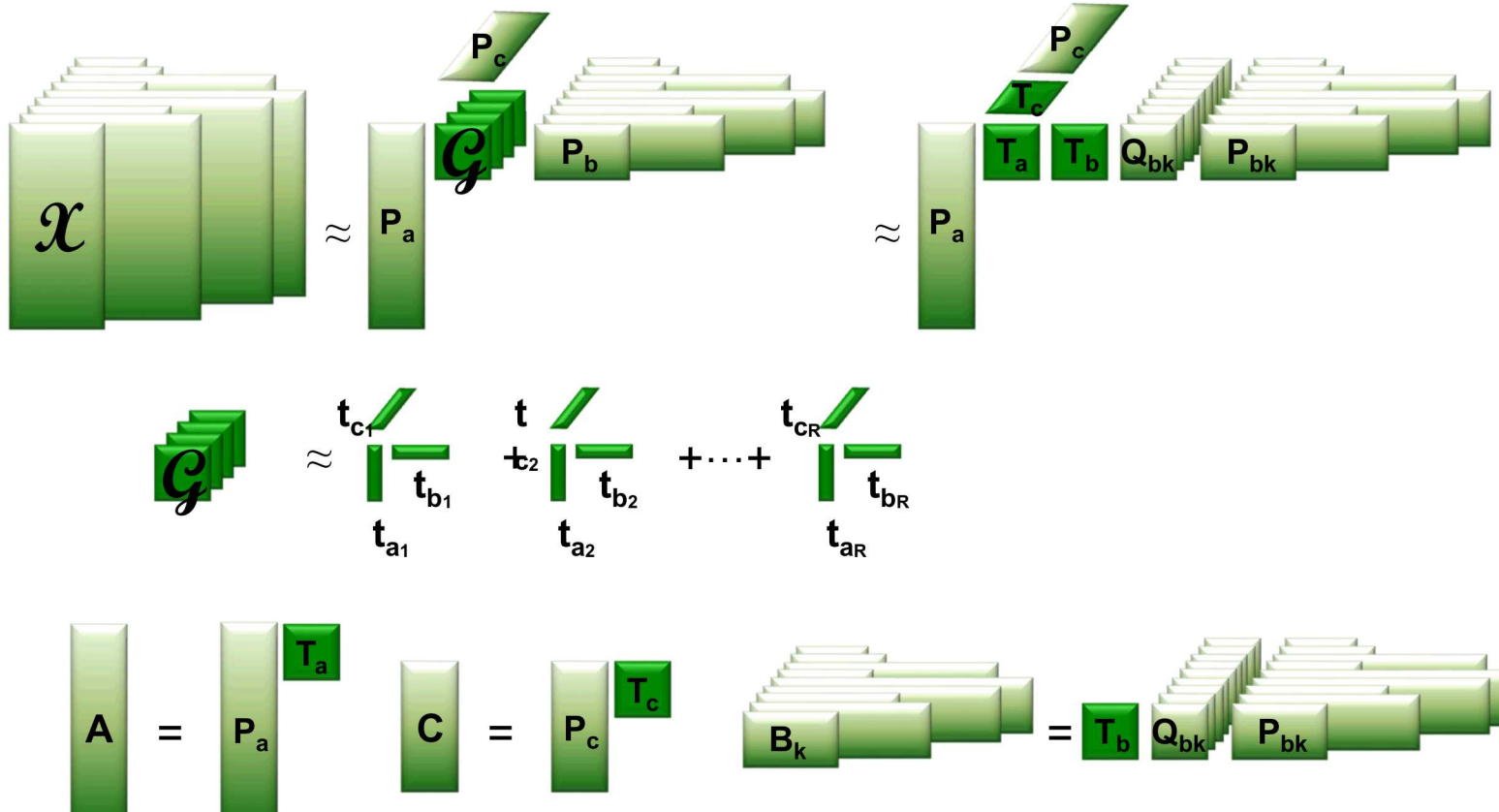
Tensor Factorization-PARAFAC2



- Data are initially individually factored
  - Orthogonal factors are retained and updated from PARAFAC on factored data



Tensor Factorization-Core PARAFAC2



PARAFAC2 Algorithms

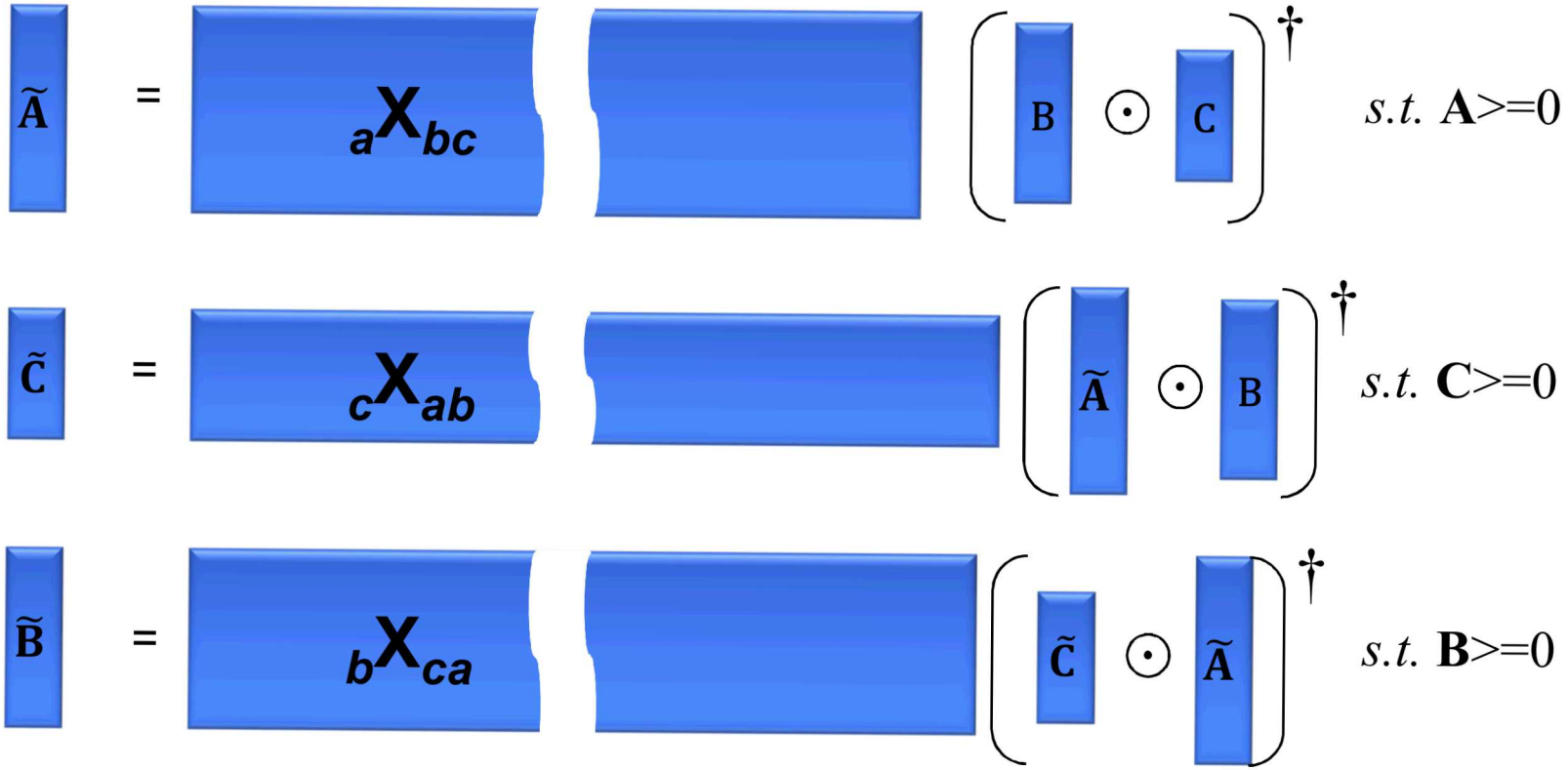
PARAFAC2

- Step 1: Initialize **A** as the eigenanalysis solution of $\mathbf{A}\Lambda = \left(\sum_k \mathbf{X}_k^{\mathrm{T}}\mathbf{X}\right)\mathbf{A}$. Initialize $\mathbf{T_b}$ and $C_1, \ldots, C_k$ as $\mathbf{I}_R$. Where the diagonal of C_k is the k^{th} column of **C**.
- Step 1a. Compute the SVD $\mathbf{T_b} C_k \mathbf{A}^{\mathrm{T}} \mathbf{X}_k^{\mathrm{T}} = \mathbf{U}_k \Delta_k \mathbf{V}_k^{\mathrm{T}}$ and update $\mathbf{P}_{\mathbf{b}k}$ as $\mathbf{V}_k \mathbf{U}_k^{\mathrm{T}}$.
- Step 1b. Update $\mathbf{T_b}$, **A** and $C_1, \ldots, C_k$ by PARAFAC algorithm applied to the three-way array **G**, with frontal planes $\mathbf{P}_{\mathbf{b}k}^{\mathrm{T}} \mathbf{X}_k$.
- Step 1c. Evaluate the residual function value $\sigma_3 = \sum_k \left\| \mathbf{X}_k - \mathbf{P}_{\mathbf{b}k} \mathbf{T_b} C_k \mathbf{A}^{\mathrm{T}} \right\|^2$. If $\sigma_3^{old} - \sigma_3^{new} > \varepsilon \sigma_3^{old}$ for some small value ε, go to Step 1a.

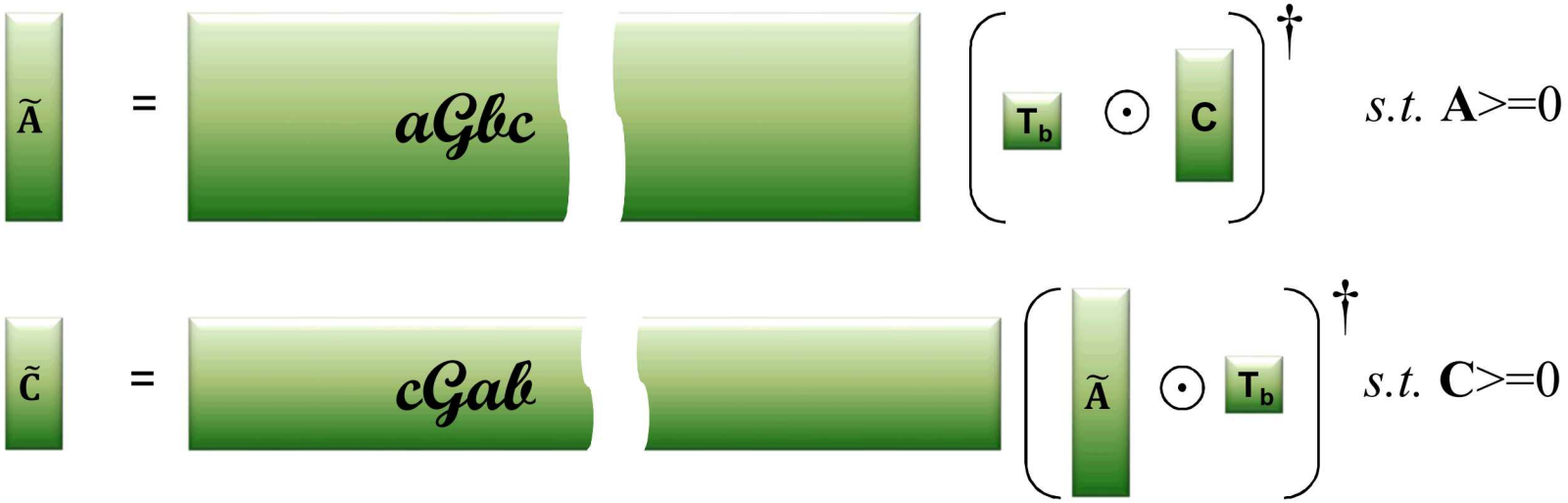
Core-PARAFAC2

- Step 1: Compute $\mathbf{P_a}$ as the eigenanalysis solution of $\mathbf{P_a}\Lambda = \left(\sum_k \mathbf{X}_k^{\mathrm{T}}\mathbf{X}\right)\mathbf{P_a}$. Initialize $\mathbf{T_a}$ and $\mathbf{T_b}$ as $\mathbf{I}_R$, and $C_1, \ldots, C_k$ as $\mathbf{I}_R$ plus $\mathbf{N}_R$. The diagonal of C_k is the k^{th} column of **C**.
- Step 1a. Compute the SVD $\mathbf{P}_{\mathrm{a}}^{\mathrm{T}} \mathbf{X}_k^{\mathrm{T}} = \mathbf{U}_k \Delta_k \mathbf{V}_k^{\mathrm{T}}$ and update $\mathbf{P}_{\mathbf{b}k}$ as $\mathbf{V}_k$, and let $\mathbf{Y}_k = \mathbf{U}_k \Delta_k$
- Step 1b. Compute the SVD $\mathbf{T_b} C_k \mathbf{T}_{\mathrm{a}}^{\mathrm{T}} \mathbf{Y}_k^{\mathrm{T}} = \mathbf{U}_k \Delta_k \mathbf{V}_k^{\mathrm{T}}$, update $\mathbf{Q}_{\mathbf{b}k}$ as $\mathbf{V}_k \mathbf{U}_k^{\mathrm{T}}$, and let $\mathbf{Z}_k = \mathbf{Y}_k \mathbf{Q}_{\mathbf{b}k}$.
- Step 1c. Compute the SVD $[\mathbf{Z}_1(:) \quad \cdots \quad \mathbf{Z}_k(:)] = \mathbf{U}\Delta\mathbf{V}$ and update $\mathbf{P_c}$ as **V** and update three-way array **G** as $\mathbf{U}\Delta$. Set $\mathbf{T_c} = \mathbf{P}_{\mathrm{c}}^{\mathrm{T}} \mathbf{C}$.
- Step 1d. Update $\mathbf{T_a}$, the $\mathbf{T_b}$, and $\mathbf{T_c}$ by one cycle of a PARAFAC algorithm applied to **G**.
- Step 1e. Evaluate the residual function value $\sigma_3 = \left\| \mathbf{G} - \otimes\left(\mathbf{T_a}, \mathbf{T_b}, \mathbf{T_c}\right) \right\|^2$. If $\sigma_3^{old} - \sigma_3^{new} > \varepsilon \sigma_3^{old}$ for some small value ε, go to Step 1a.

Nonnegativity-PARAFAC

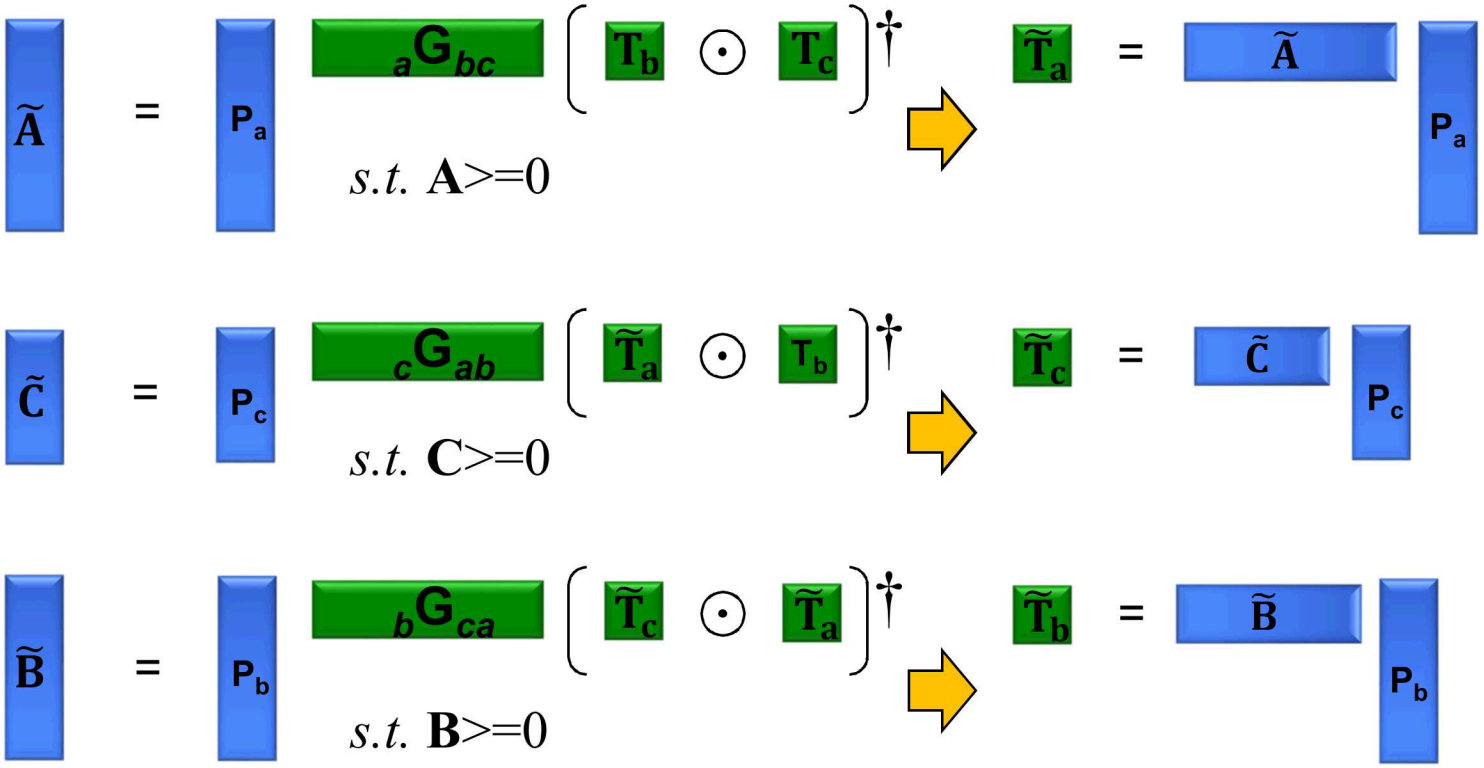


Nonnegativity-PARAFAC2



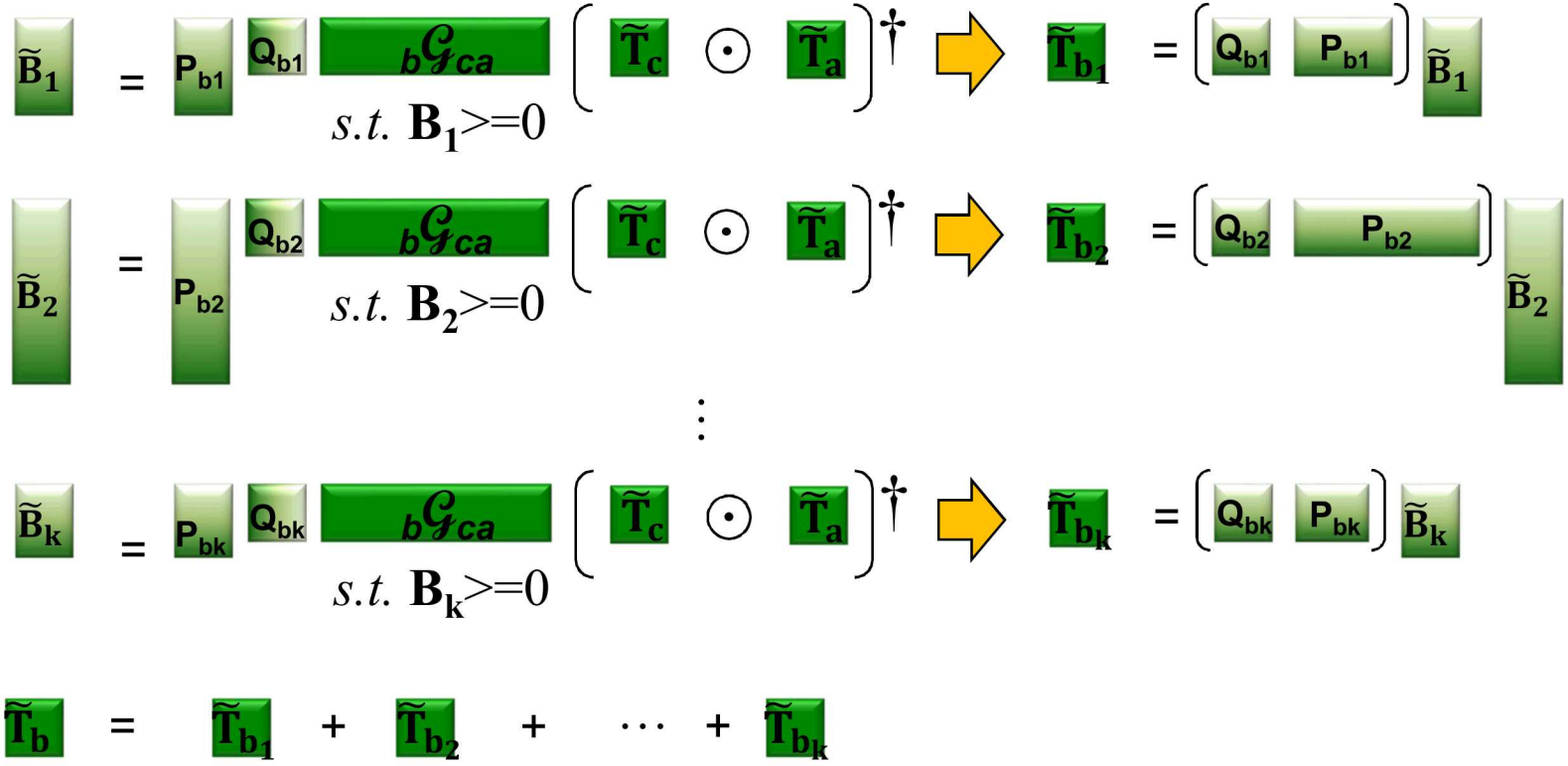
No mechanism for B-mode nonnegativity.
(New Flexible PARAFAC2 describes coupling method)

Nonnegativity-Core PARAFAC



1) Solve for the factors in a rigorous NNLS sense
2) Project nonnegative factors into orthogonal factor space
3) Yields rotation matrix for ~nonnegative factors

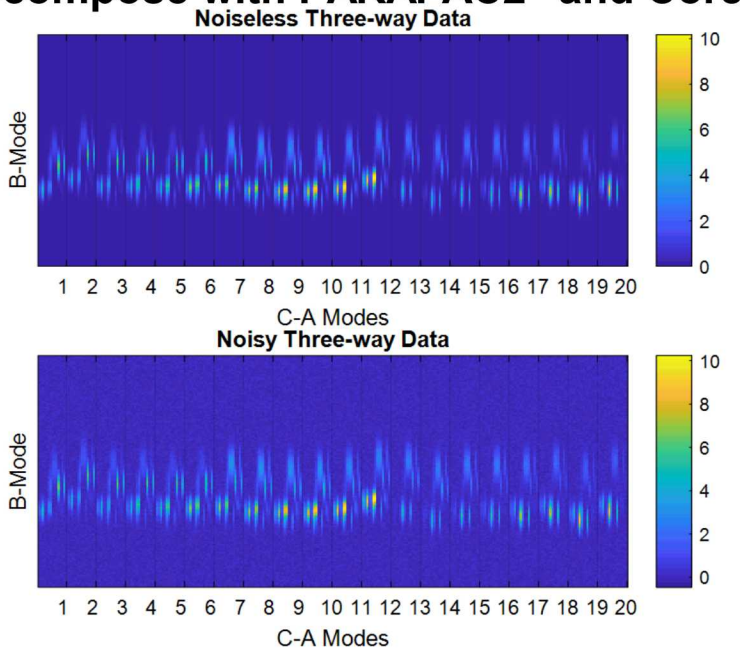
Nonnegativity-Core PARAFAC2

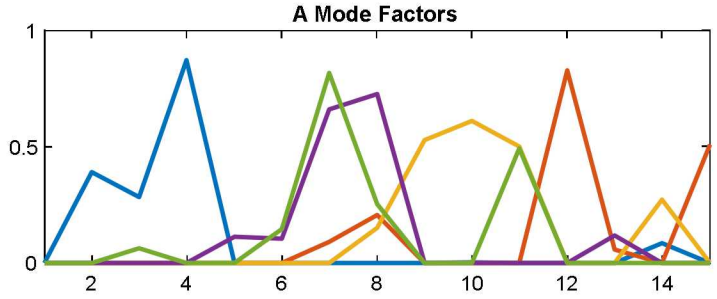


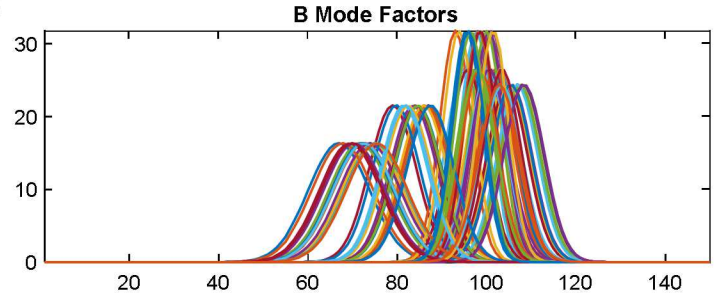
1) Solve for A and C modes rotation matrices as in NN-Core PARAFAC
2) For B-mode, solve for each layer factors in a rigorous NNLS sense
2) Project nonnegative factors into orthogonal factor space for each layer
3) Add rotation matrices for each layer to get overall B-mode rotation matrix

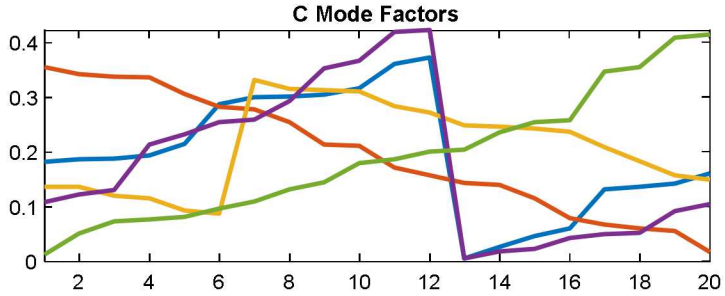
Synthetic Data

- **Data parameters:**
  - **Dimensions: 15 x 150 x 20 (A x B x C)**
  - **Rank 5**
  - **Shifts in Gaussian-shaped B-mode factors**
- **Gaussian Noise**
  - **Added to ~100 PSNR**
- **Decompose with PARAFAC2* and Core-PARAFAC2**









* https://www.mathworks.com/matlabcentral/mlc-downloads/downloads/submissions/1089/versions/1/download/zip/parafac2.zip (downloaded 5/5/18)

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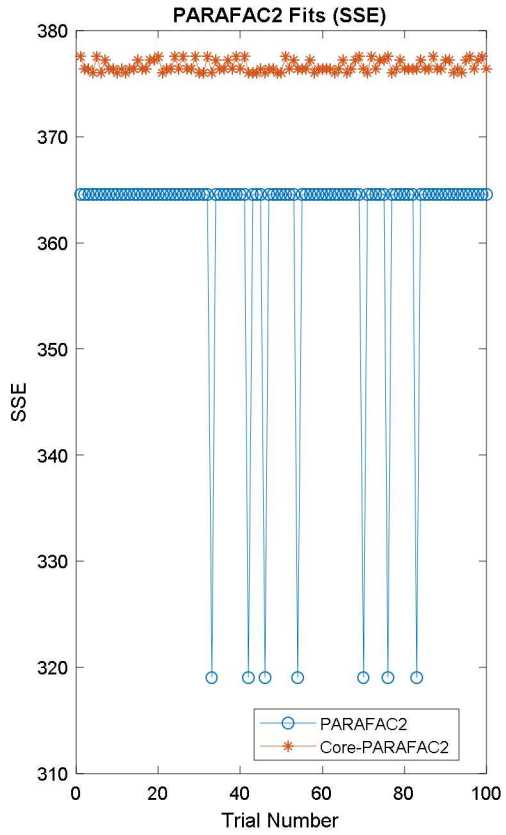
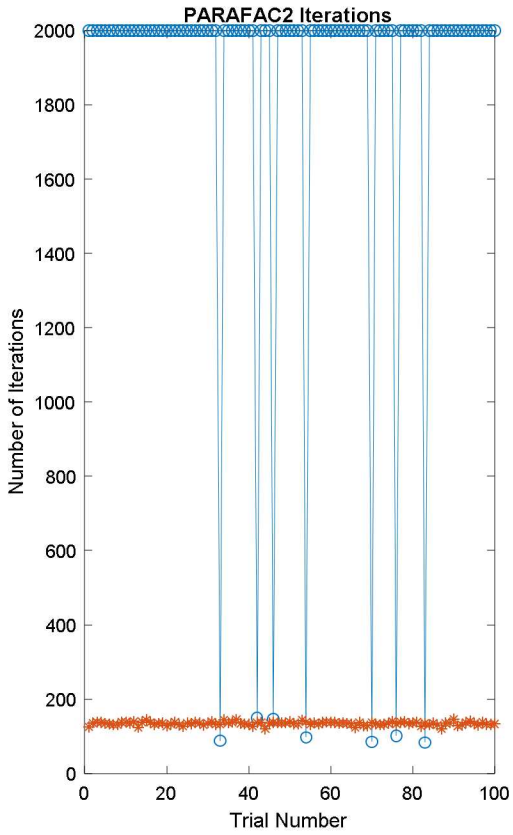
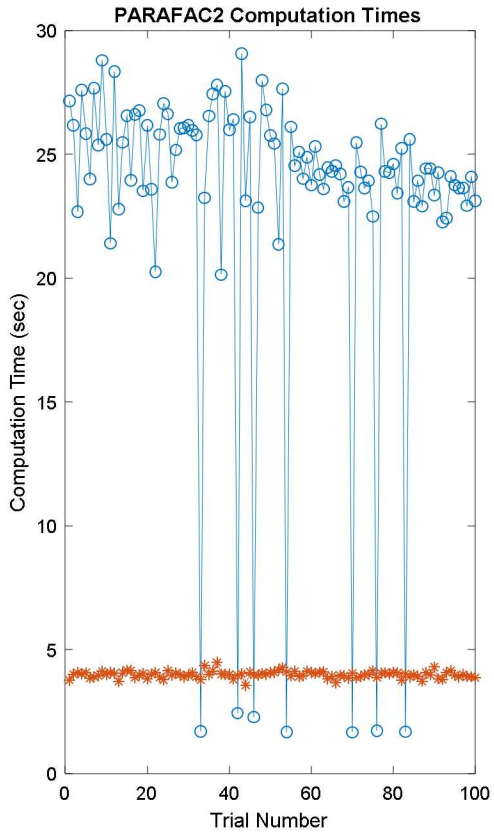
PARAFA2 and Core PARAFAC2 Fitting Statistics







Core-PARAFAC2 appears to have much more computational stability
Standard PARAFAC2 produced "good" fits ~7% of the time.

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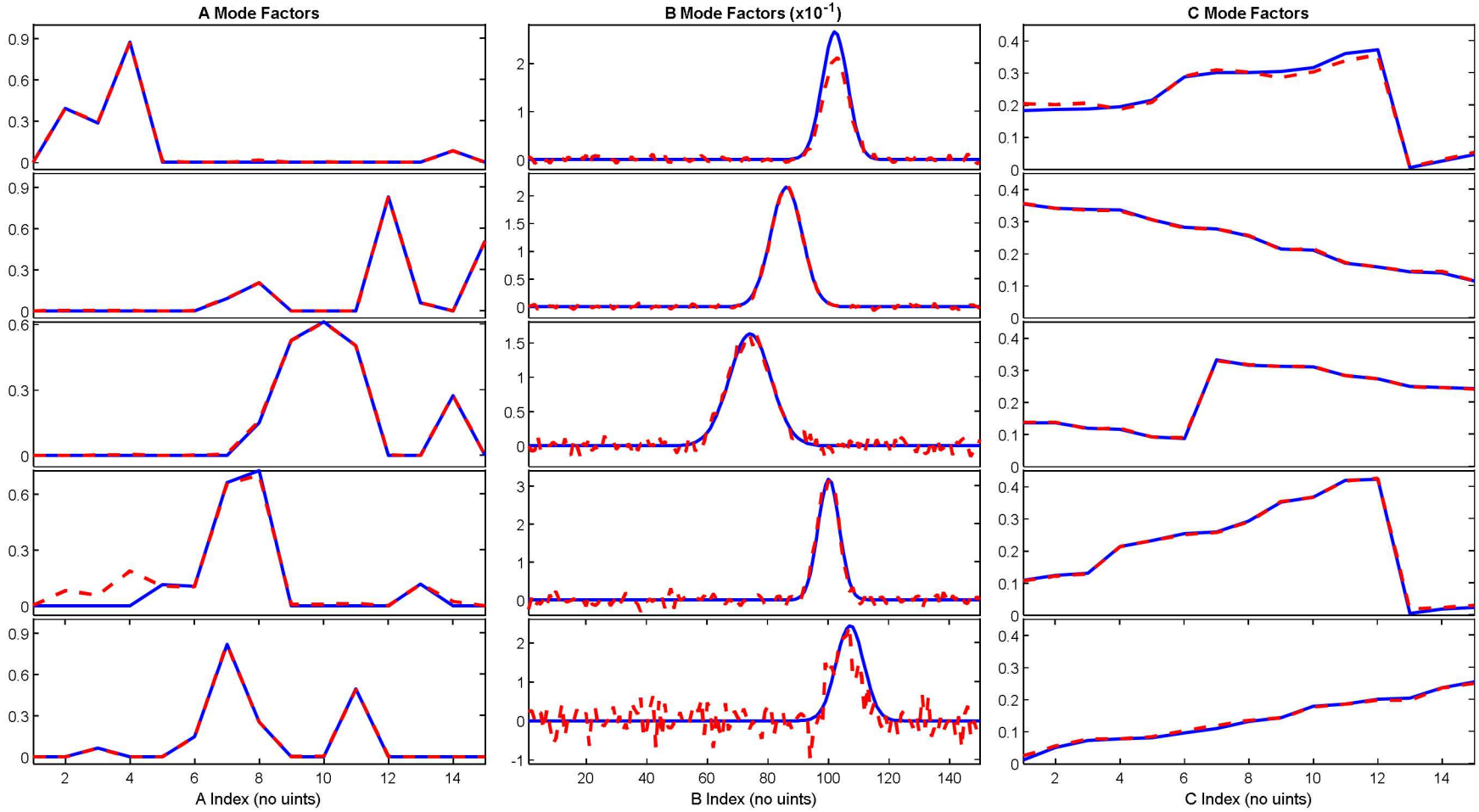
PARAFAC2 Best Fit (first slab)



PARAFAC2 does a nice job on this synthetic data set, some of the time.

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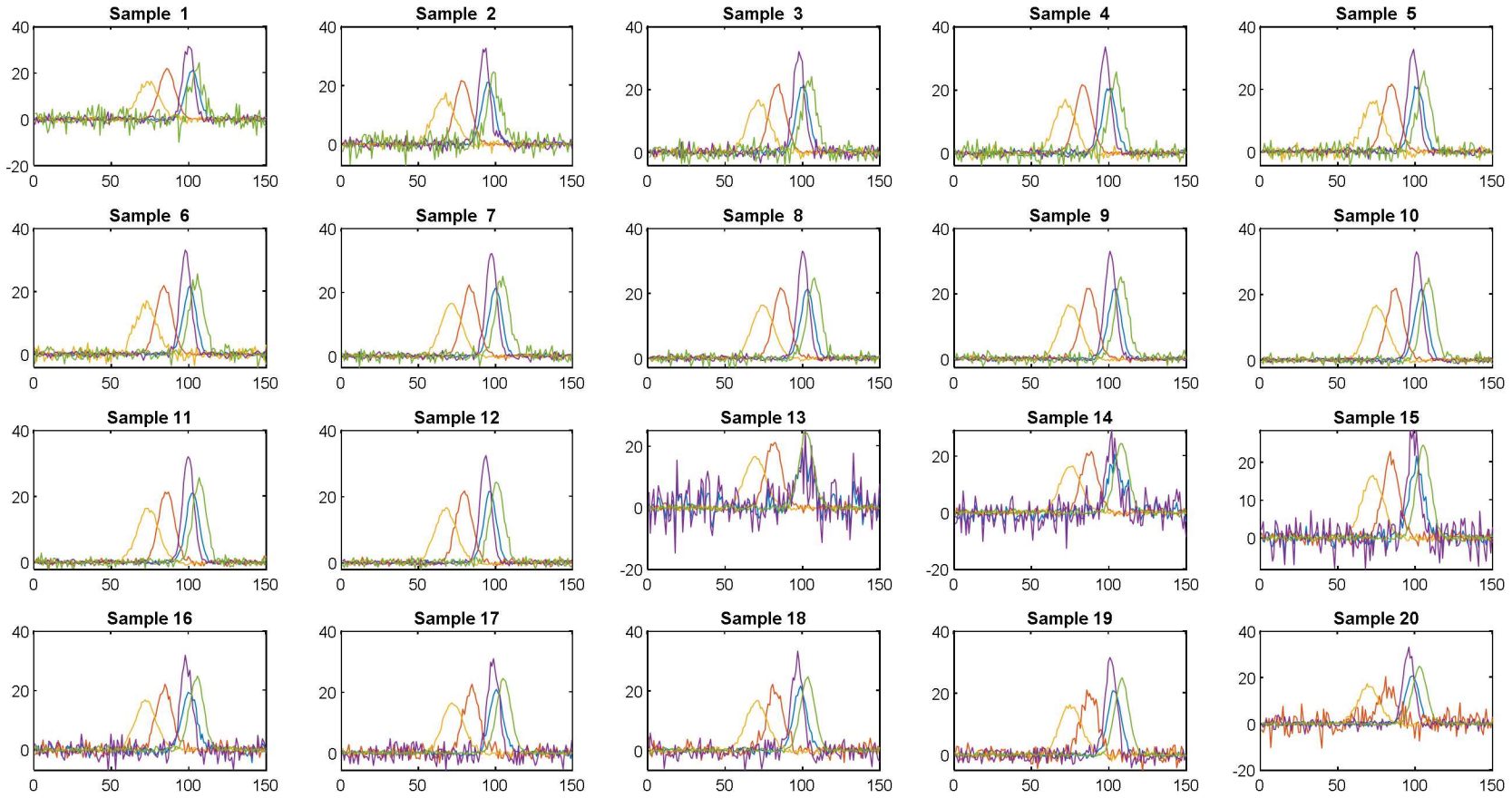
PARAFAC2 B-Mode Best Fit (all slabs)



Does not require nonnegativity constraints, but many trials.

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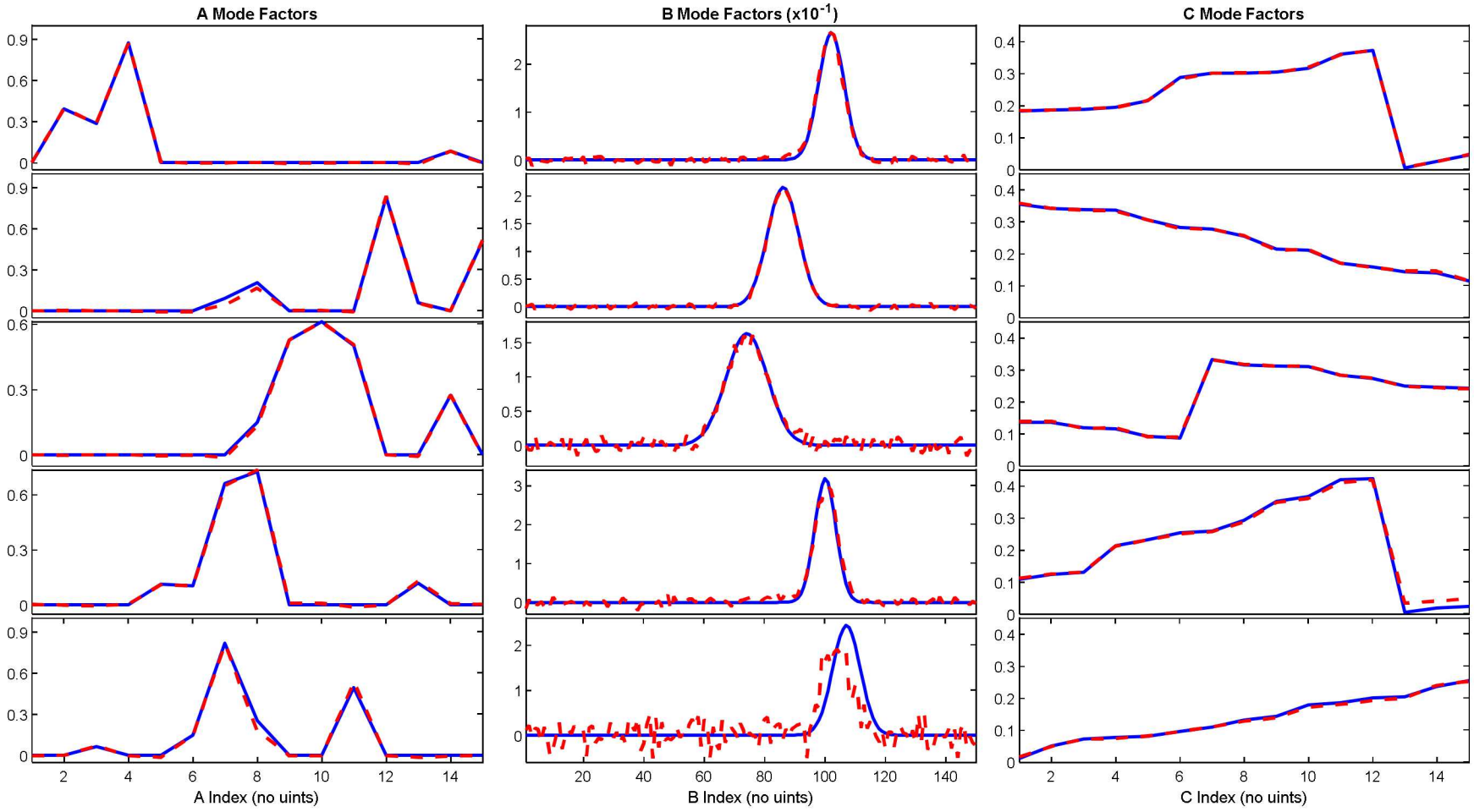
Core PARAFAC2 Best Fit (first slab)



Core PARAFAC2 also does a nice job on this synthetic data set.

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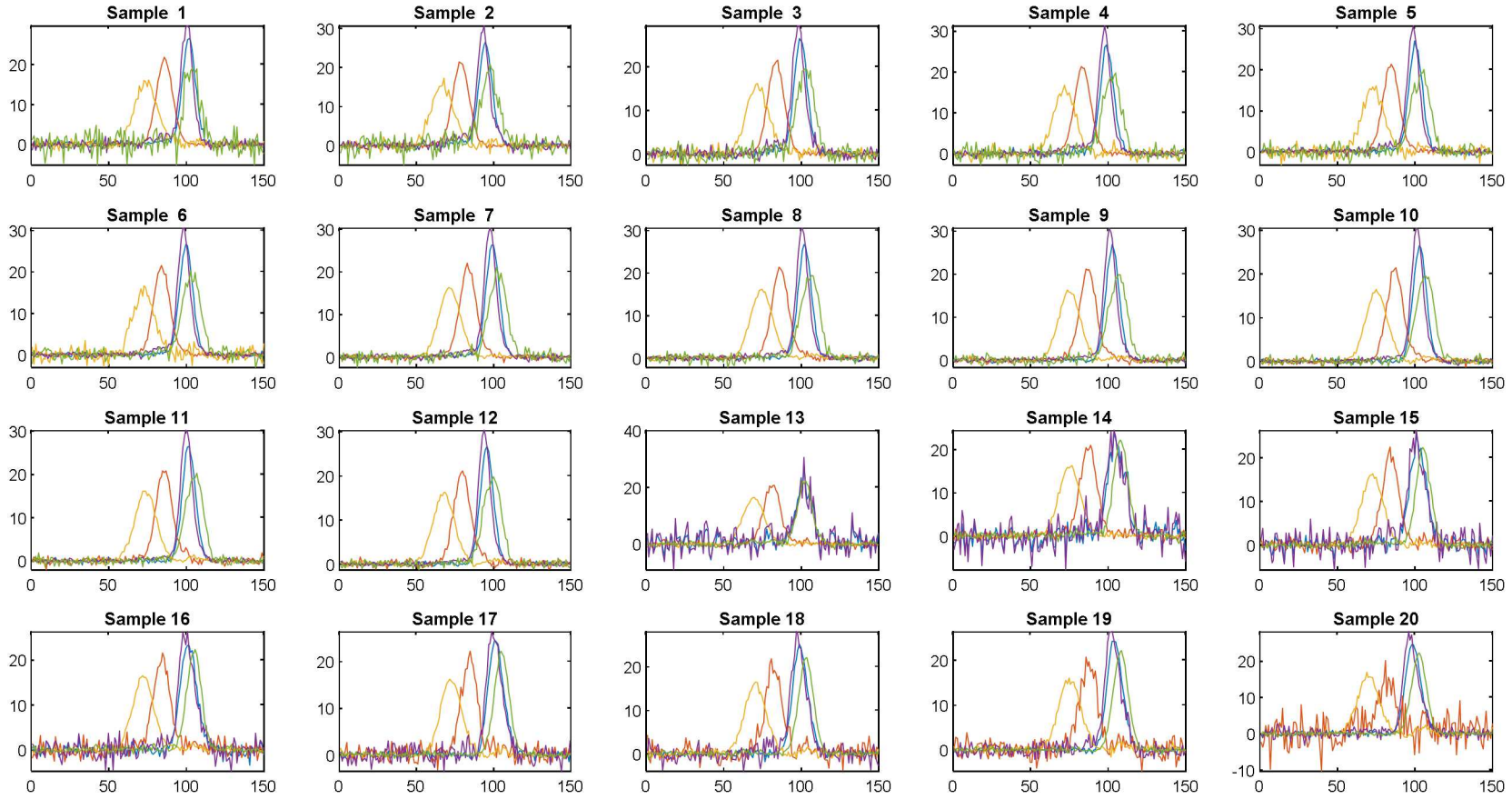
Core PARAFAC2 B-Mode Best Fit



Imposing nonnegativity constraints gives consistent results.

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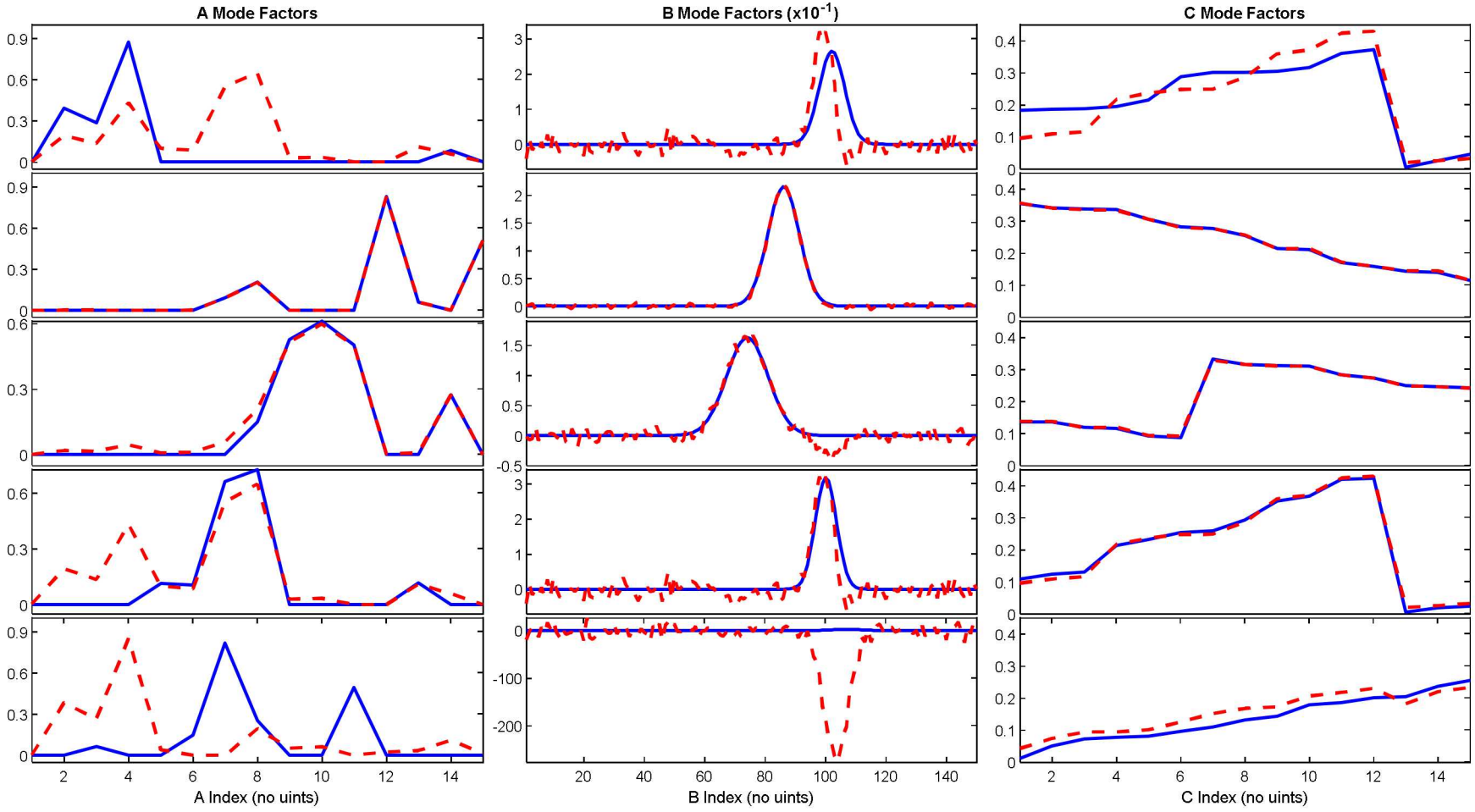
PARAFAC2 Worst Fit



PARAFAC2 sometimes produces suboptimal results.

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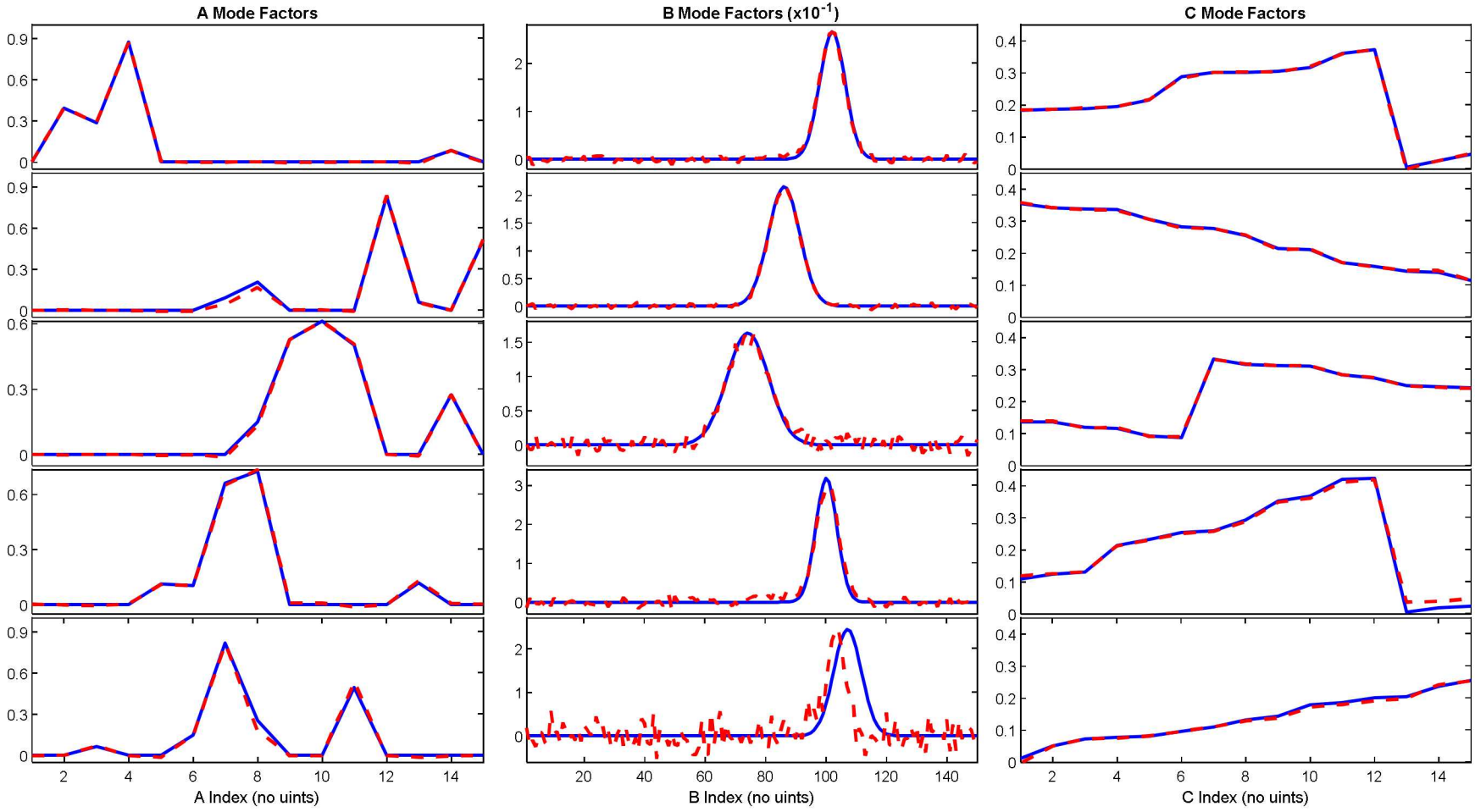
Core PARAFAC2 Worst Fit



Core PARAFAC2 with nonnegativity gives stable results.

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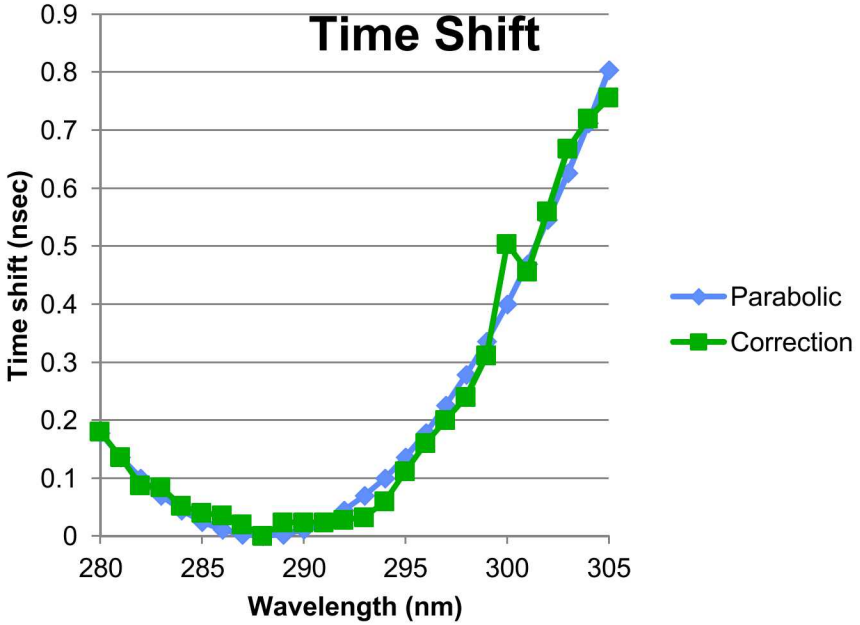
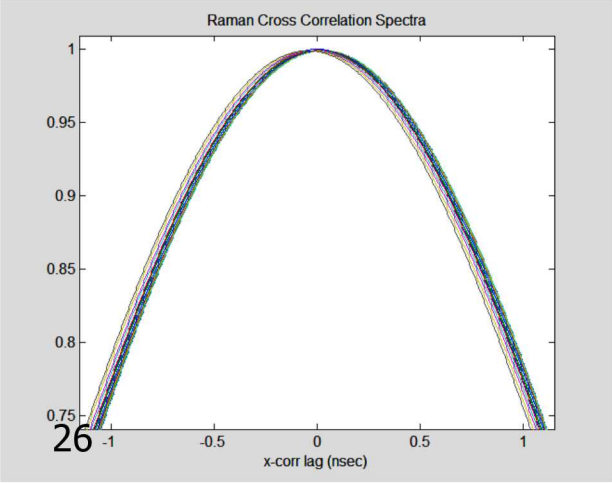
Experimental Data

- Solution of six EPA priority pollutants in methanol
  - Chemical Species:
    - Fluorene, Naphthalene, Pyrenene, Phenanthrene, Benzo[k]fluoranthrene, Benzo[b]fluoranthrene
    - From Sigma-Aldrich used without further purification
    - Solutions prepared in HPLC grade methanol.
- Collect a single TREEM
  - Original data size:
    - 640 time-mode elements
    - 251 emission wavelength-mode elements
    - 26 excitation wavelength-mode elements
- Acquisition
  - Collect waveforms: ~130 nsec duration in 200 psec increments
  - Step emission wavelengths: 300 – 550 nm in 1 nm increments
  - Excitation wavelengths: 280 – 305 nm in 1 nm increments
  - Collect each WTM in approximately 5 minutes

Correction of Time Domain Shifts

- Due to the laser gain profile, there is an excitation wavelength dependent shift in the time domain
  - Shift is approximately parabolic with wavelength
- Correction involves estimating shift with Raman or Rayleigh scattered excitation
  - Interpolate profiles at 50x measured data
  - Find shift values using cross-correlation
- Correct TREEM data
  - Interpolate profiles at 50x measured
  - Shift data requisite intervals
  - Down-sample data
  - Size after: 636 elements





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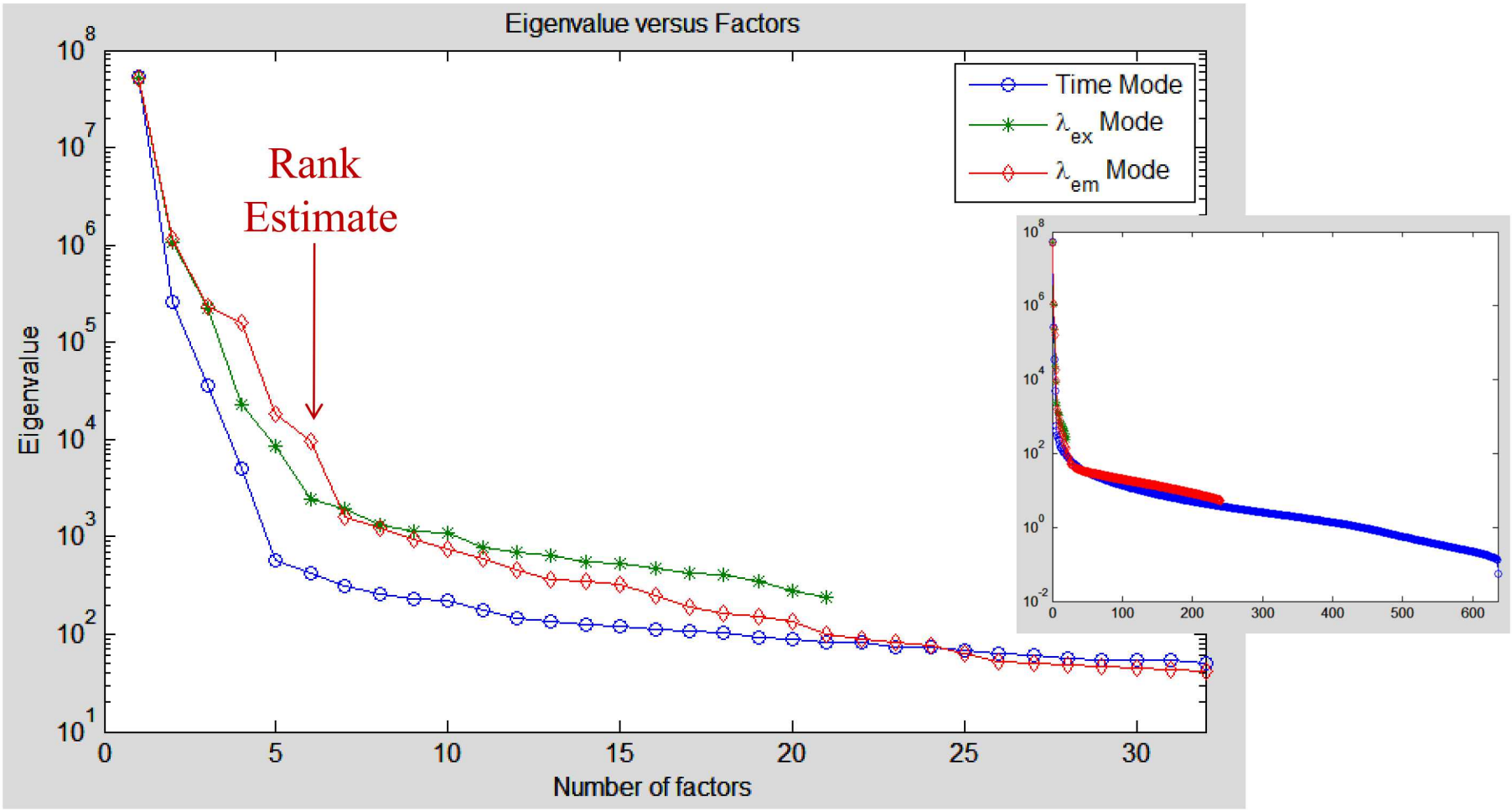
Data Analysis

- Wavelength ranges, λ_{ex}: 280-300 nm, λ_{em}: 310-550 nm
- PARAFAC on shift-corrected data
  - Use fast Tucker1 based PARAFAC algorithm
  - Initialize with random factors, 6 factor model
  - Employ fast rigorous nonnegative least squares
  - Data size: 636×241×21
- PARAFAC2
  - Utilize code from Bro as published[1]
  - Initialize with default, "best of 10 runs…", 100 separate trials
  - Impose nonnegativity in excitation & emission modes
  - Data size 640×241×21
- Core-PARAFAC2
  - Utilize code developed at SNL
  - Initialize with random factor matrices, 100 separate trials
  - Impose nonnegativity in excitation, emission & temporal modes
  - Data size 640×241×21
- Computations were performed using MATLAB®
  - Dell Precision T7910 workstation, with two, 14-core, 2.6 GHz Intel® Xeon (ES-2697) processors and 192 Gbyte RAM

1. Kiers, H.A.L. TenBerge, J.M.F. Bro, R. J. Chemom., 13 (1999) 275-294.

Eigenvalue Plot



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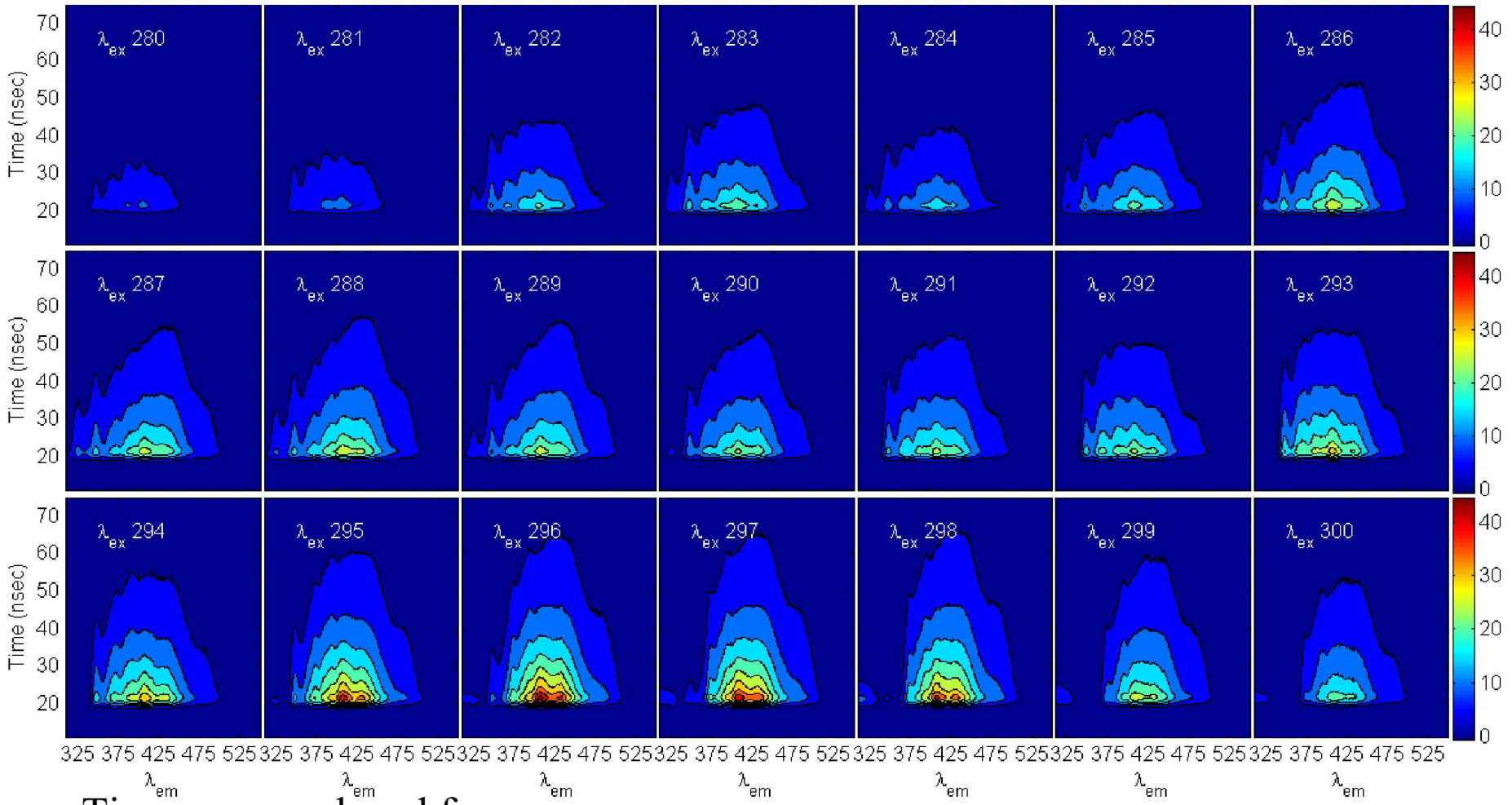
Raw TREEM Data as 21 WTMs



Time range reduced for improved comparison.

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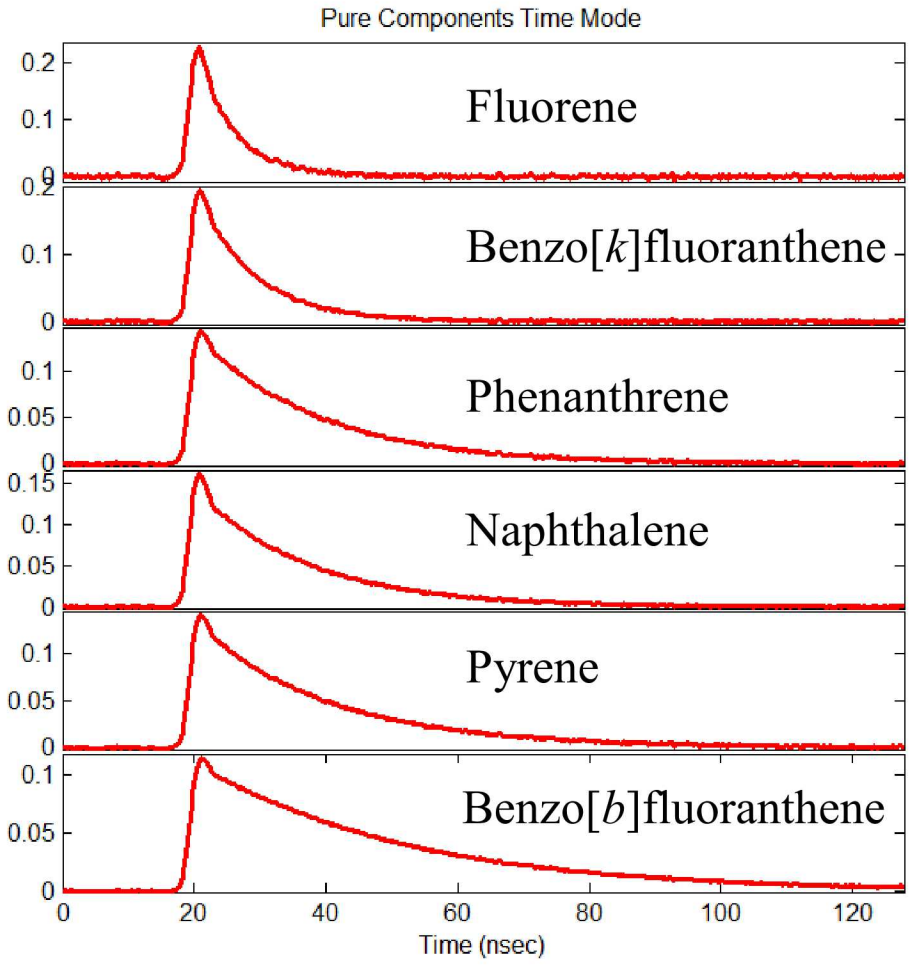
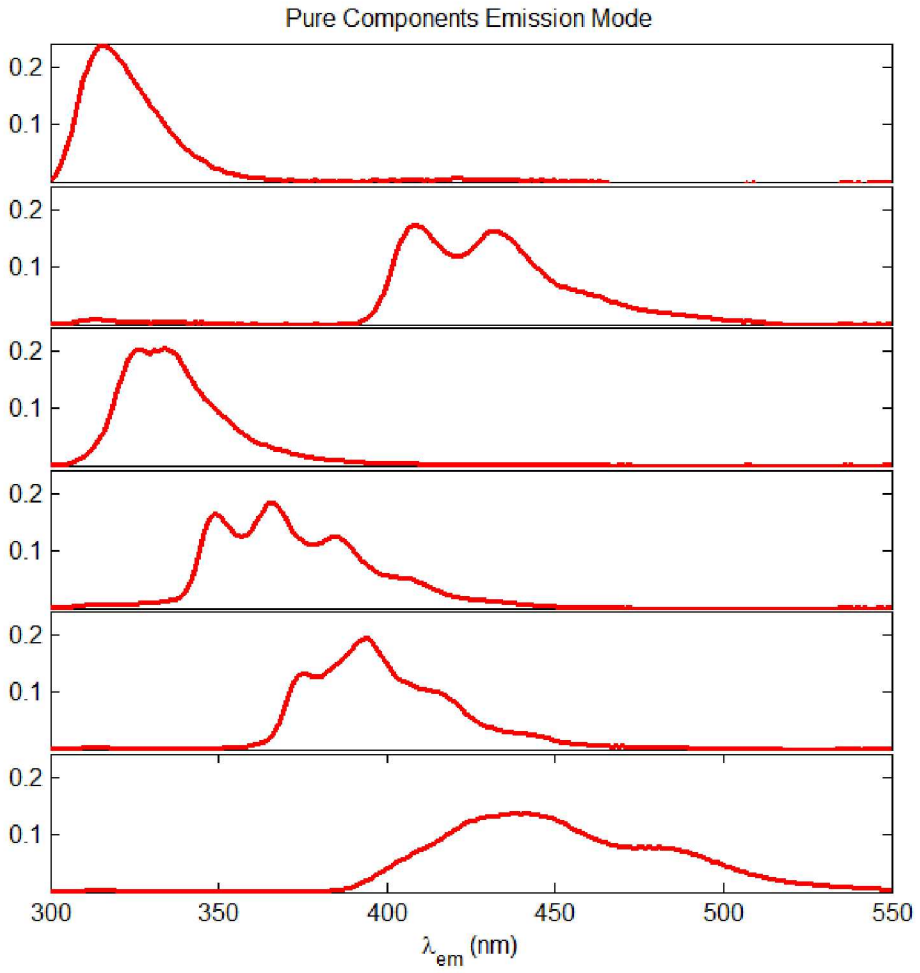
Pure Component Time and Emission Modes





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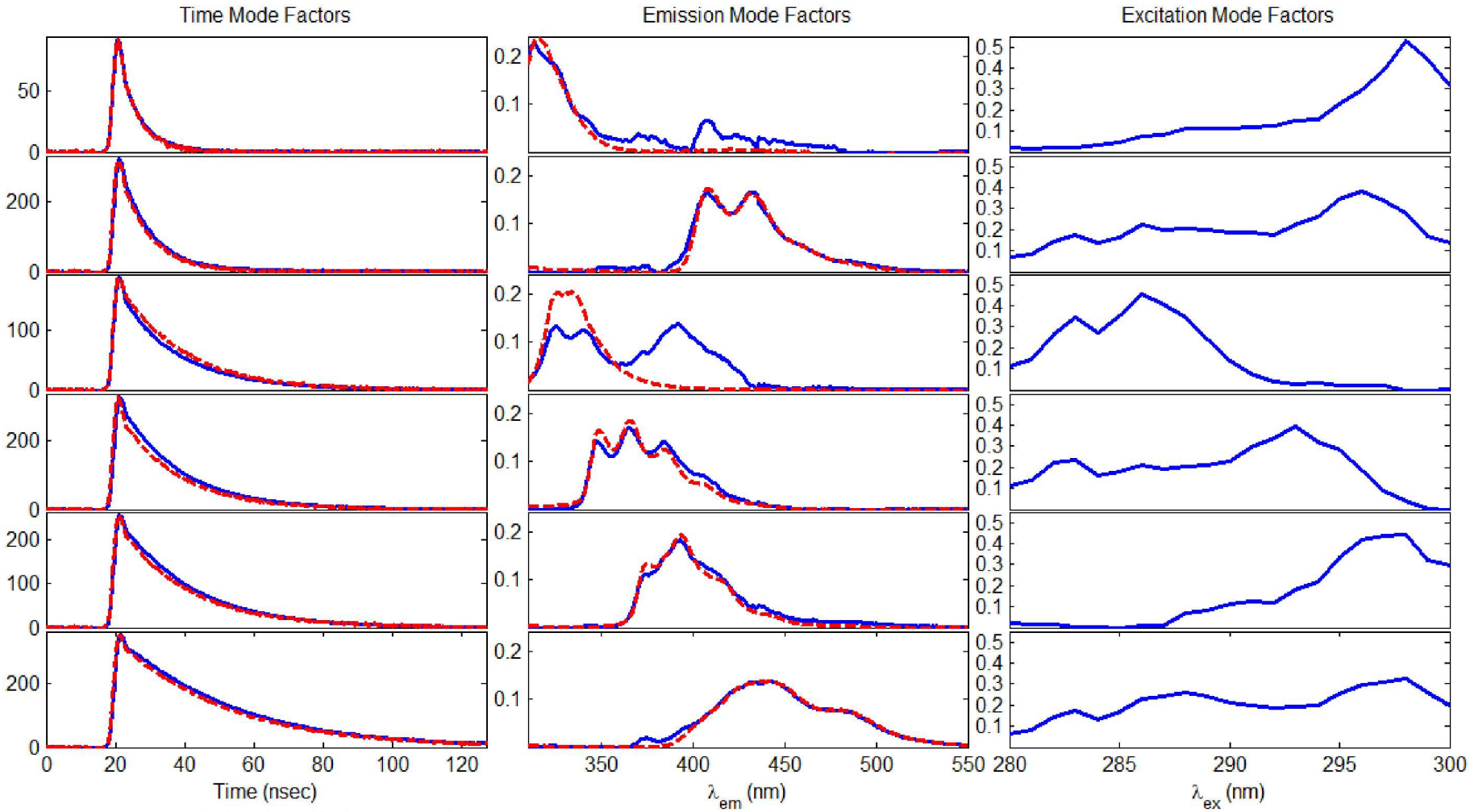
Six Factor Fast-PARAFAC Model



Nonnegativity in all three modes.
Computation time: 14 seconds
Blue: Fast PARAFAC, Red: Pure Components

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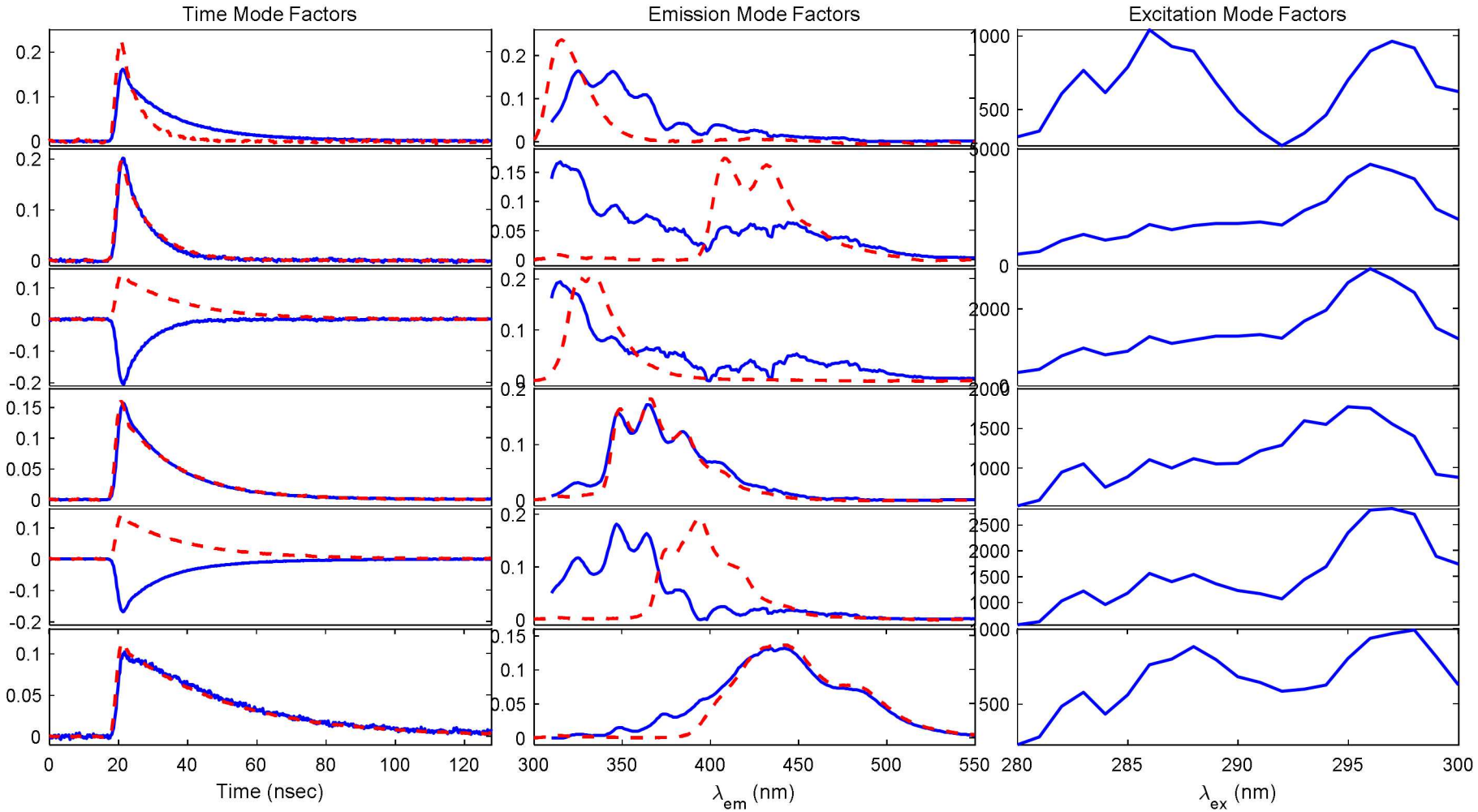
Six Factor PARAFAC2 Model Best Fit



Nonnegativity in Emission and Excitation modes.
Computation time: 357 seconds
Blue: PARAFAC2, Red: Pure Components

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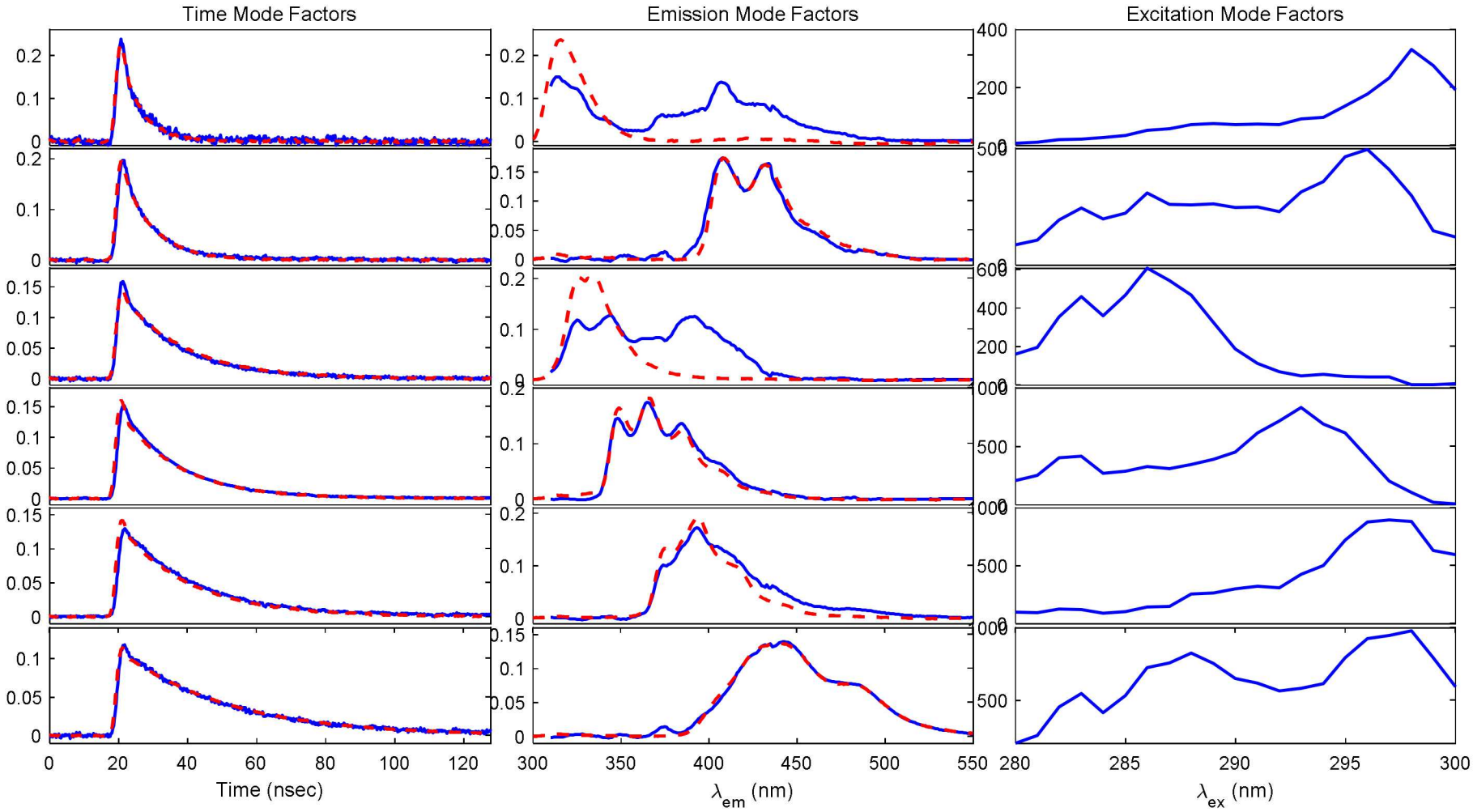
Six Factor Core-PARAFAC2 Model Best Fit



Nonnegativity in all three modes.
Computation time: 147 seconds
Blue: Core-PARAFAC2, Red: Pure Components

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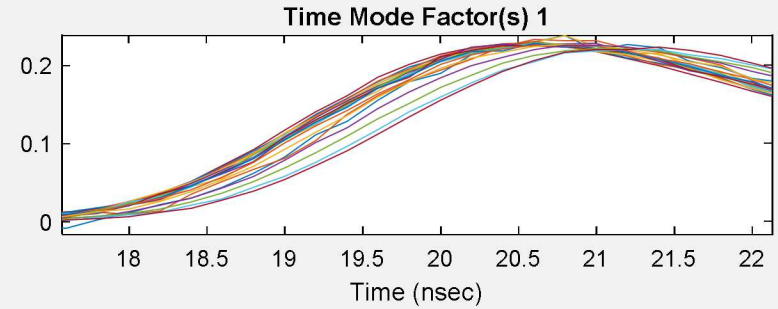
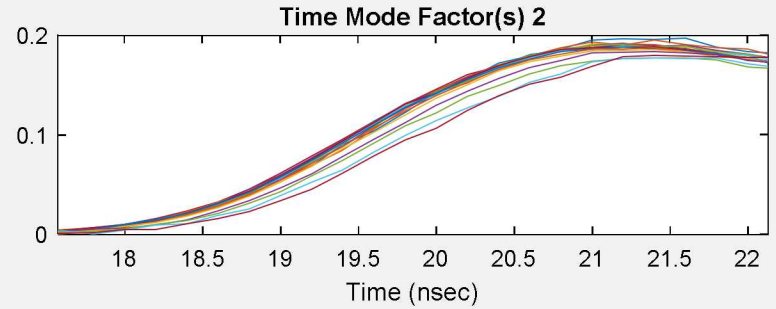
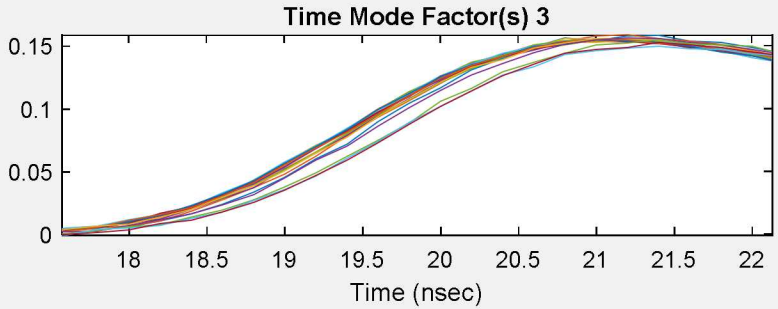
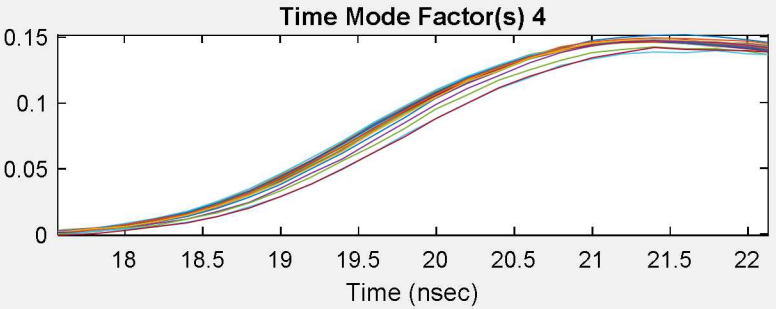
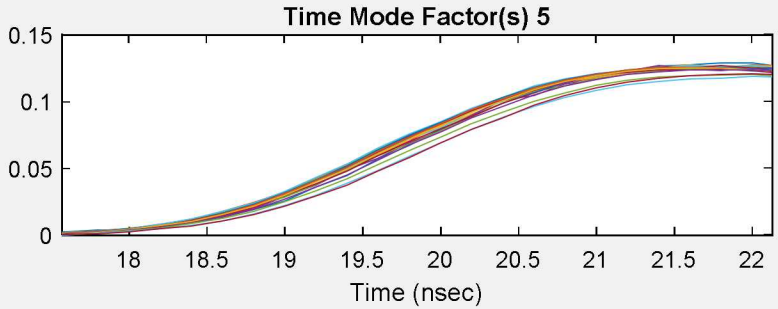
All Time (shifted) Mode Factors

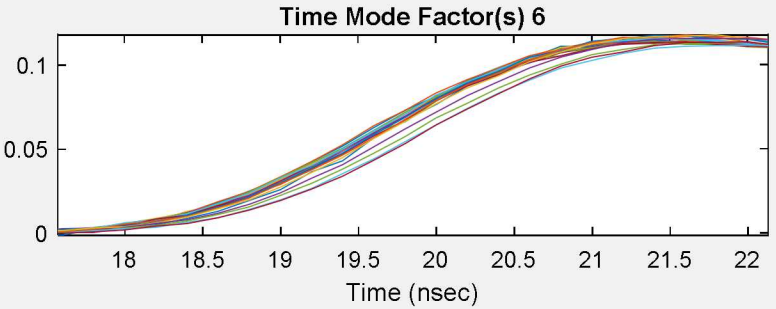




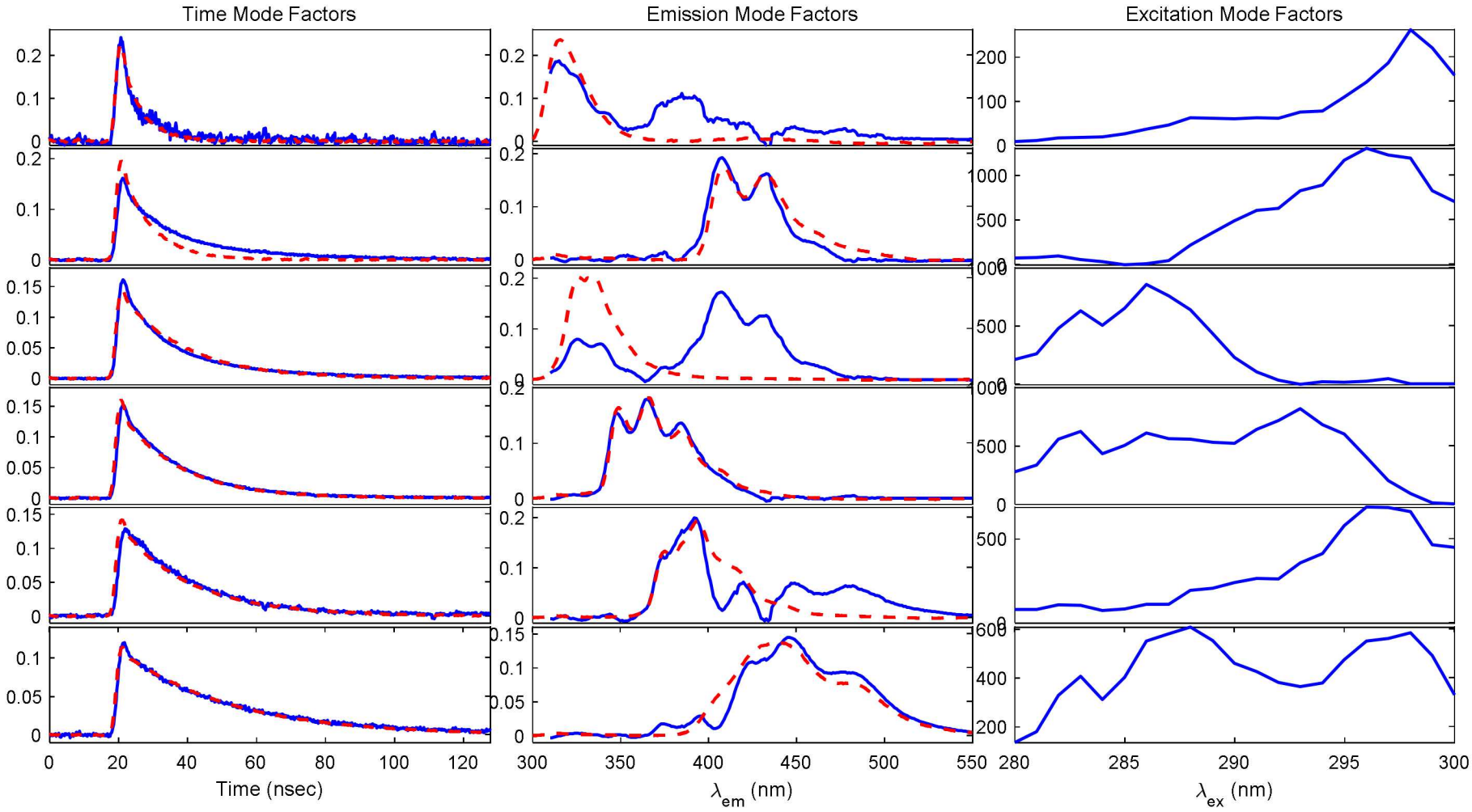








Six Factor Core-PARAFAC2 Model Worst Fit



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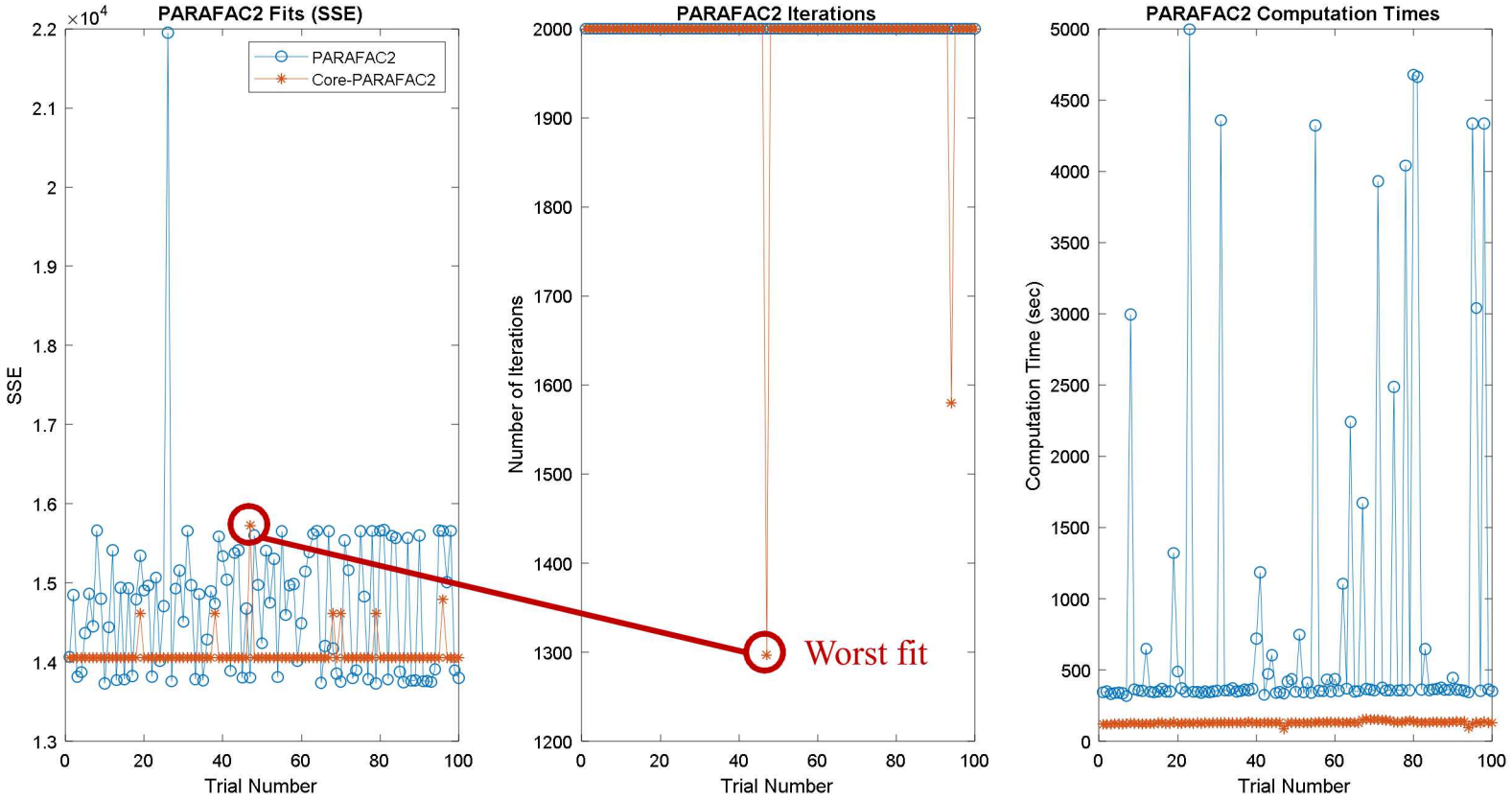
PARAFA2 and Core PARAFAC2 Fitting Statistics



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Results and Conclusions

- Core PARAFAC2 with nonnegativity, under appropriate circumstances, provides excellent, stable factor analysis
- Fast PARAFAC with imposed nonnegativity renders reasonable pure component factors for most species
  - Time shifting algorithm appears to make a suitable correction for follow-on analysis
  - Predicted pure component emission spectra and decay curves are in good agreement with single species solution data.
- PARAFAC2 is appropriate for time-shifted data, but inability to impose nonnegativity in time domain results in poor factor resolution
- Core PARAFAC2 with nonnegativity constraints provides results highly comparable to the shift-corrected data
  - Demonstrated excellent results in both the simulated and real data

Future Work

- Investigate methods to “oversample” core array
  - Using factor ranks greater than model rank
  - Easily done in Core PARAFAC
- Determine method to optimize number of PARAFAC cycles utilized on core
- Compare Core PARAFAC2 with other new methods

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