New multi-way models and algorithms for solving blind source separation problems

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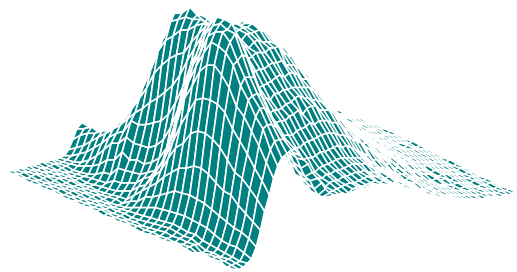


Content

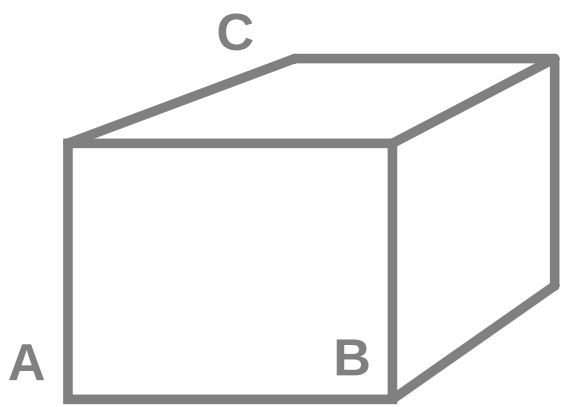
- Introduction
- Models & algorithms
- Applications
- Problems

Three-way data?

- Simply a set of two-way matrices
- Each mode consist of same basic entities over the other modes
- E.g. same samples measured at different variables, several times
- Instead of a matrix with typical elements x_{ij}, we have an array with elements x_{ijk}

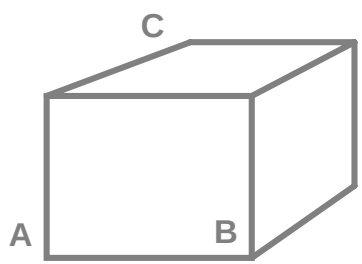
Where?

- Sensory analysis, Process analysis, Image analysis, Experimental design, Spectroscopy, Chromatography, Environmental analysis, QSAR, Communication, Medicine, ...



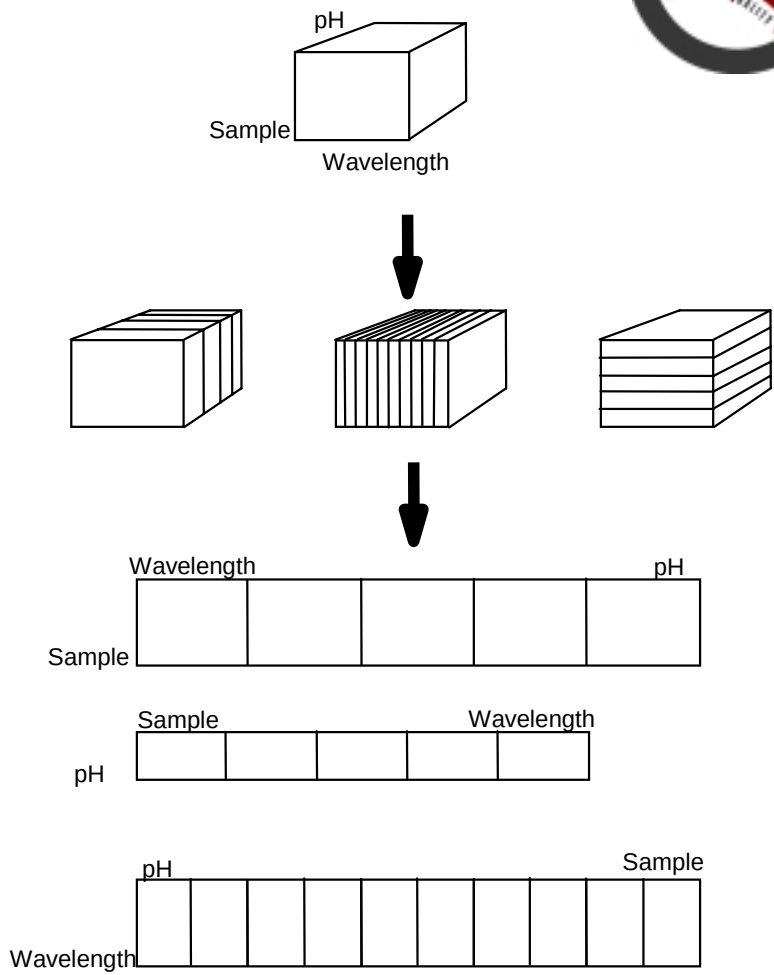
Examples from chemistry

- **Sensory analysis**
  - Score as a function of (Food sample, Judge, Attribute)
- **Process analysis**
  - Measurement as a function of (Batch, Variable, time)
  - Measurement as a function of (Variable, Lag, Location)
- **Image analysis**
  - Pixelvalue as a function of (Sample, Image pixel, Variable)
- **Experimental design**
  - Response as a function of (factor 1, factor2, factor3,..)
- **Spectroscopy**
  - Intensity as a function of (Wavelength, Retention, Sample, Time, Location , Treatment)
- **Environmental analysis**
  - Measurement as a function of (Location, Time, Variable)



Unfolding/matricization

- Traditional approach
  - Unfolding leading to two-way data and analysis
- Three-way models
  - Natural extensions of two-way models
  - PCA leads to PARAFAC or Tucker3 depending on how it is extended
  - Rank-reduced regression, e.g. PLS leads to multilinear PLS (N-PLS)



Unfolding/matricization

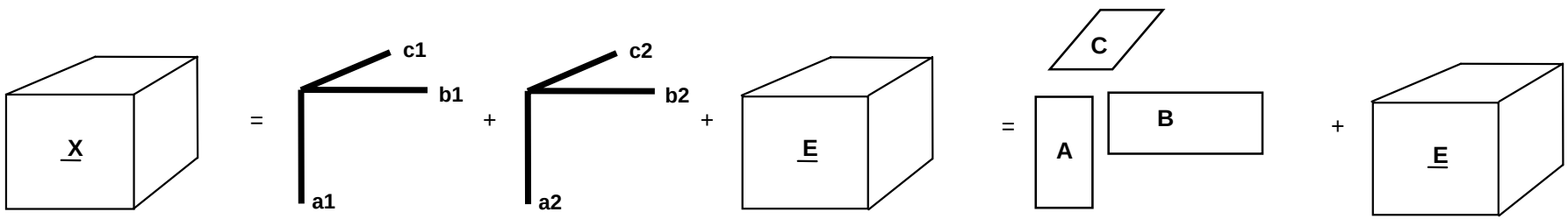
Often leads to overfitting because nature of model ≠ nature of the data

Unfold: *Wold, Geladi, Esbensen, Öhman. Principal component- and PLS-analyses generalized to multi-way (multi-order) data arrays. Copenhagen Symposium on Applied Statistics:249-277, 1986.*

PARallel FACtor analysis

- PCA - bilinear model,
  - $x_{ij} = \sum_f t_{if} p_{jf} + e_{ij}, \quad i=1,..,I;\ j=1,..,J$

- PARAFAC - trilinear model,
  - $x_{ijk} = \sum_f a_{if} b_{jf} c_{kf} + e_{ijk}, \quad i=1,..,I;\ j=1,..,J;\ k=1,..,K$

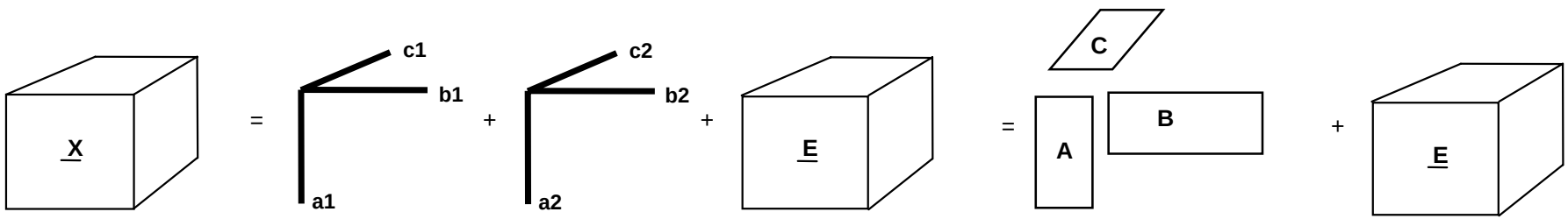


PARallel FACtor analysis

- PCA - bilinear model,
  - $\mathbf{X} = \mathbf{AB}' + \mathbf{E}$

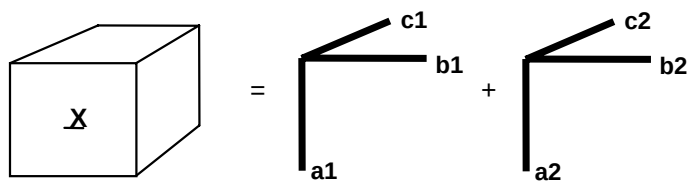
- PARAFAC - trilinear model,
  - $\mathbf{X}_k = \mathbf{AD}_k\mathbf{B}' + \mathbf{E}_k$, $k = 1,\ldots,K$

    $\mathbf{D}_k = \mathrm{diag}(\mathbf{C}(k,:))$



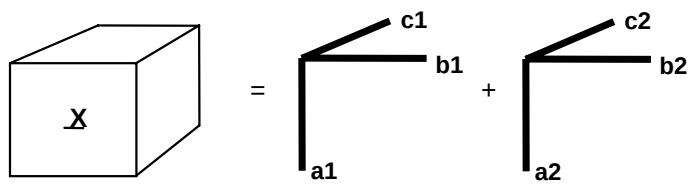
Definition of three-way rank

- Rank of two-way matrix
  - Minimum number of bilinear (PCA) components needed to reproduce matrix
- Rank of three-way array
  - Minimum number of trilinear (PARAFAC) components needed to reproduce array



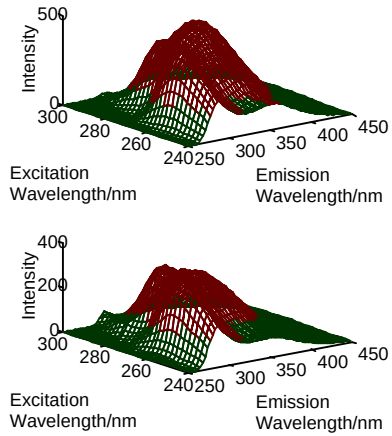
Definition of three-way rank

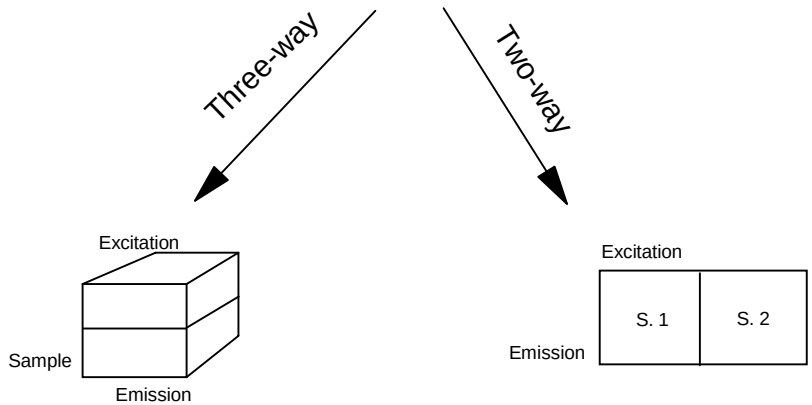
- Two-way
  - Any random matrix has full rank with probability one
  - Row-rank = column-rank
  - Simple rules for maximal rank
- Three-way
  - Random 2×2×2 is rank 2 (30%) and rank 3 (70%)!
  - Row-rank ≠ column-rank ≠ tube-rank!
  - No rules for maximal rank!
    - Except simple cases such as 3×3×3 is five.



PARAFAC - uniqueness

- PARAFAC has a unique property: It is unique!!
  - Practical example: Fluorescence
  - Two samples
  - Mixture of three analytes
    - (Trp, Tyr, Phe)





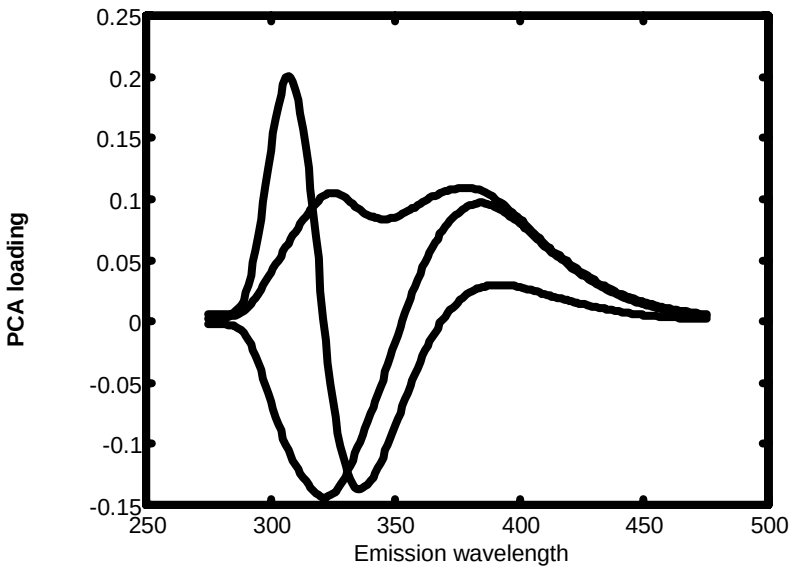
PARAFAC invented in 1970 by Harshman and independently by Carroll & Chang under the name CANDECOMP. Based on a principle of parallel proportional profiles suggested in 1944 by Cattell

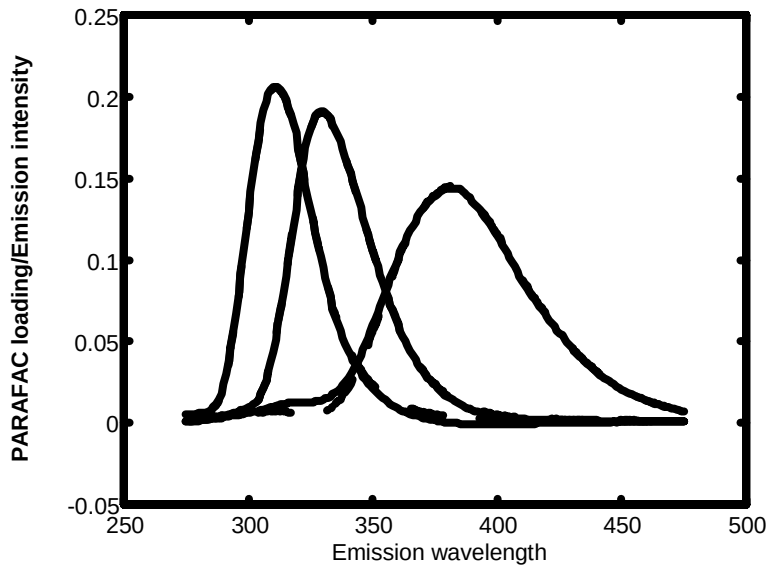
•R. A. Harshman. *UCLA working papers in phonetics* 16:1-84, 1970.
•J. D. Carroll and J. Chang. *Psychometrika* 35:283-319, 1970.
•R. B. Cattell. *Psychometrika* 9:267-283, 1944.

PARAFAC - uniqueness

- PCA orthogonal loadings

PARAFAC pure spectra!





PARAFAC for CDMA

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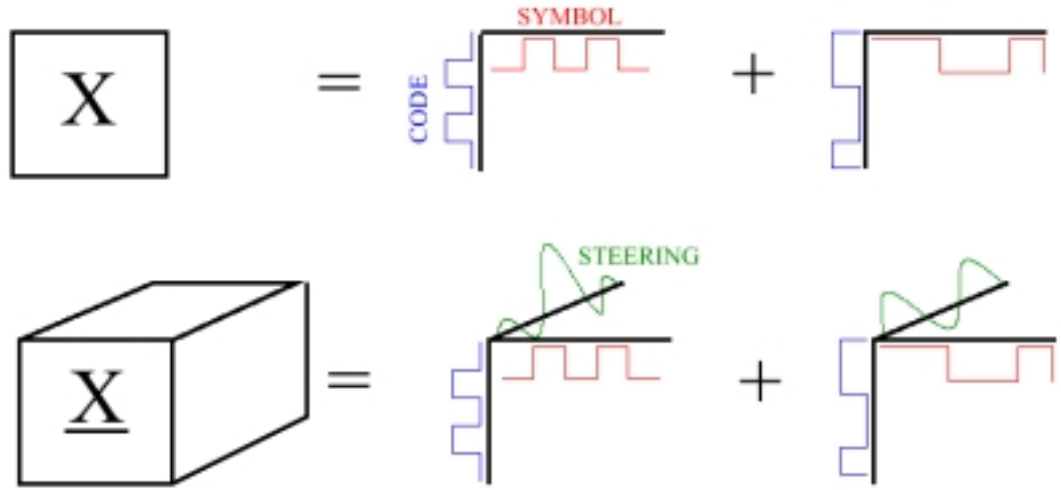
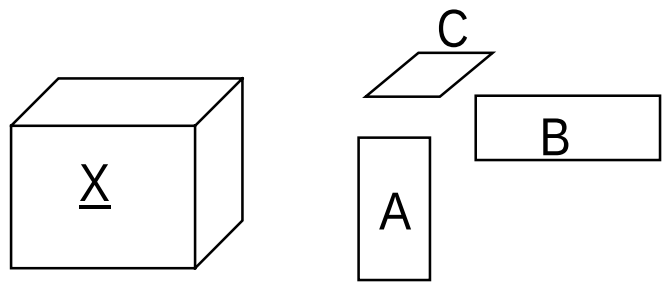
Blind CDMA



- Fact 1: Low-rank matrix (2-way array) decomposition not unique
- Fact 2: Low-rank 3- and higher-way array decomposition (PARAFAC) is unique

Uniqueness - conditions

A PARAFAC model is unique when



$$k_A + k_B + k_C \geq 2F + 2$$

F is the number of components and k_A is the k-rank of loading **A** = maximal number of randomly chosen columns which will have full rank ($\leq F$)

J. B. Kruskal. *Linear Algebra and its Applications* 18:95-138, 1977.
SidGiaBro, IEEE TSP, Mar 2000, $N = 3$, $\mathbb{R}$
SidBro. *JChemom* 14 2000, any N, $\mathbb{C}$ $\sum_{n=1}^{N} k_n \geq 2F + (N-1)$

PARAFAC - uniqueness

- **Uniqueness - what does it mean?**
  - Mixtures of analytes can be separated
  - Concentrations can be estimated
  - Pure spectra and profiles can be estimated
    - $8 \times 8 \times 8$ array is unique for a 10 component model!!
    - Hence with 8 snapshots, 8 symbols and 8 codes, recovery of 10 sources is possible
- **Examples**
  - On-line purity control (Kodak) Windig, *J. Magnetic Resonance* 132:298-306, 1998
  - Determination of kinetic parameters Bijlsma, *AIChE Journal* 44:2713-2723, 1998
  - Mathematical chromatography Leurgans, *Statistical Science* 7:289-319, 1992
  - Localizing cellular phone calls Sidiropoulos & Bro. PARAFAC Techniques for Signal separation. In: *Signal Processing Advances in Communications*, edited by P. Stoica, G. B. Giannakis, Y. Hua, and L. Tong,Prentice-Hall, 2000,

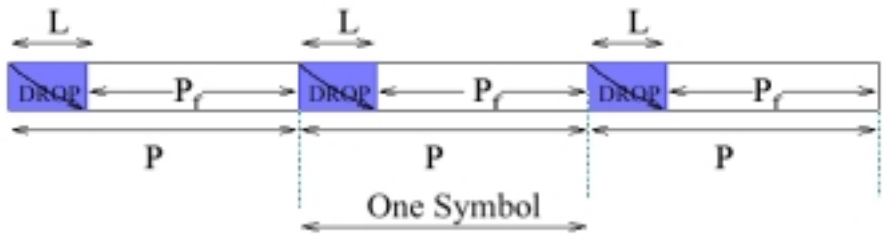
Significance

- All codes, steering unknown; no need for training
- LS (cond. ML) $\rightarrow$ symbols, codes, steering for all users
- Iterative, $O(FIJK)$
- Diversity combining: non $\perp$ codes, $F > K, F > I, F > J$
- Deterministic, modulation-independent
- Frequency-selective multipath? Performance?

PARAFAC - uniqueness

Frequency-Selective Multipath

- (SS) Symbol-level (but not chip-level) synchronization
- (FF) Multipath reflectors in the far field
- (FIR) FIR channels, max order = L (incl. asynchronism)
- (DP) Spreading gain $K = K_f + L$, and $K_f \geq 2$ ISI-free chips (suffering from ICI only) are available ("discard prefix"):



e.g., [QS-CDMA-Iltis, Bensley-Aazhang, Liu-Xu, Tsatsanis et al]

PARAFAC - uniqueness

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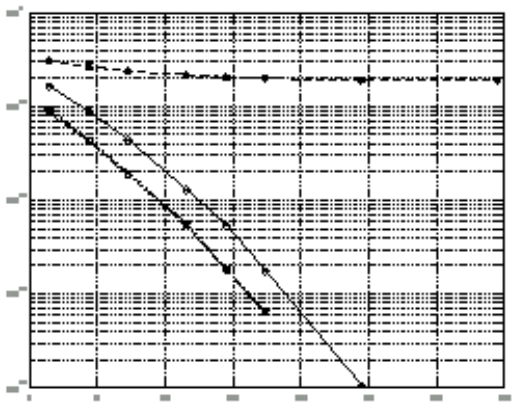
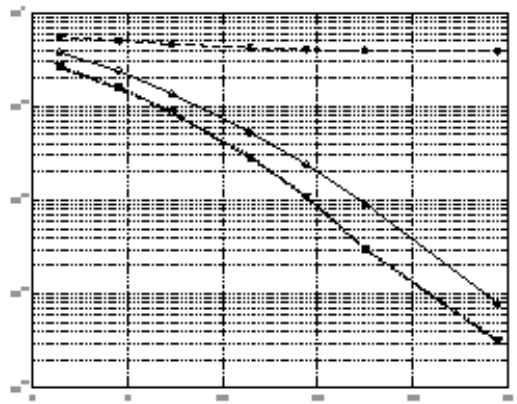
Performance: $F = 4, I = 2, J = 50, K = 6$



DE-BPSK



DE-QPSK

- Blind performance close to non-blind MMSE

Alternating least squares algorithm

1. Initialize **B** and **C**
2. $\mathbf{A} = \left(\sum_{k=1}^{K} \mathbf{X}_k \mathbf{B} \mathbf{D}_k \right) \left\{ (\mathbf{B}'\mathbf{B}) * (\mathbf{C}'\mathbf{C}) \right\}^{-1}$
2. $\mathbf{B} = \left(\sum_{k=1}^{K} \mathbf{X}'_k \mathbf{A} \mathbf{D}_k \right) \left\{ (\mathbf{A}'\mathbf{A}) * (\mathbf{C}'\mathbf{C}) \right\}^{-1}$
3. $diag\mathbf{D}_k = \left\{ (\mathbf{B}'\mathbf{B}) * (\mathbf{A}'\mathbf{A}) \right\}^{-1} \mathrm{diag}(\mathbf{A}'\mathbf{X}_k\mathbf{B}), k = 1,..,K$
4. Step 2 until relative change in fit is small

- * Hadamard (elementwise product)

PARAFAC - algorithm

- **Speed-up**
  - Compression of data (LS or Spline-based bases)
  - Line-search etc.

- **Alternative algorithms**
  - GRAM/DTLD – direct generalized eigenvalue problem but not LS
  - PMF3 – Gauss-Newton: fast but problematic for large arrays
  - COMFAC – Uses compression and separation + Gauss-Newton

Tucker3 – an extension of SVD

- PCA formulated as truncated SVD – **X** = **USV**' + **E**

- *Problem*:
  Express **X** in terms of new unitary bases **U** and **V**
- *Solution*:
  Regress **X** on **U** and **V**: **S** = $\mathbf{U^{+}XV'^{+}}$ = **U'XV**
- *Hence*:
  S equals (approximates) **X** in a new (truncated) coordinate system
- *Curiosity*:
  S is diagonal, but that's not mandatory

Tucker3/SVD

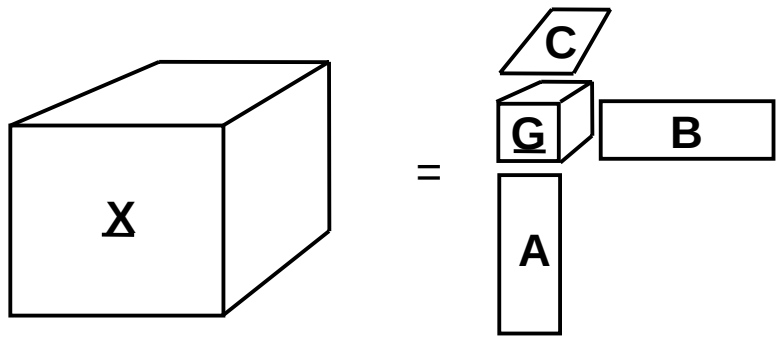
- PCA = SVD can be written (note the off-diagonals)

$$\mathbf{X} = \mathbf{A}\,\mathbf{S}\,\mathbf{B}^T = [\mathbf{a}_1 \quad \mathbf{a}_2]\begin{bmatrix} 1 & 0 \\ 0 & 2 \end{bmatrix}[\mathbf{b}_1 \quad \mathbf{b}_2]^T$$

$$= \mathbf{a}_1\mathbf{b}_1^T + 0\mathbf{a}_2\mathbf{b}_1^T + 0\mathbf{a}_1\mathbf{b}_2^T + 2\mathbf{a}_2\mathbf{b}_2^T == \mathbf{a}_1\mathbf{b}_1^T + 2\mathbf{a}_2\mathbf{b}_2^T$$

The Tucker3 model

- For three-way data, three unitary bases, **A**, **B**, and **C**; one for each mode
- Tucker3 is $\mathbf{X} = \mathbf{AG}(\mathbf{C} \otimes \mathbf{B})' + \mathbf{E}$
- Loadings are truncated bases and **G** the representation of **X** in these reduced spaces



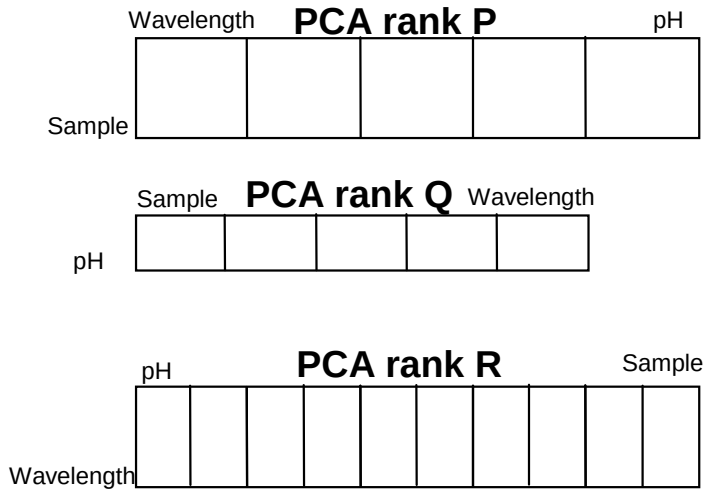
•L. R. Tucker. The extension of factor analysis to three-dimensional matrices. In: *Contributions to Mathematical Psychology*, edited by N. Frederiksen and H. Gulliksen, New York:Holt, Rinehart & Winston, 1964, p. 110-182.

•L. R. Tucker. Some mathematical notes on three-mode factor analysis. *Psychometrika* 31:279-311, 1966

Tucker3

- Important theorem
  - *If* the matricized ranks in the three modes are found to be *P*, *Q*, and *R* respectively
  - *then* a (*P*,*Q*,*R*) Tucker3 model fit the data perfect

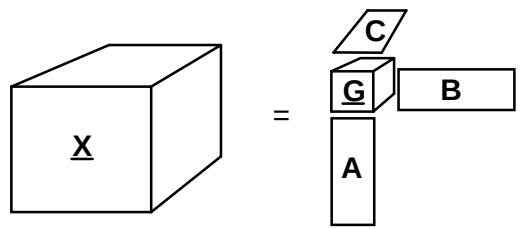
- Practical value
  - If the pseudo-ranks are definitely found to be *P*, *Q*, and *R* respectively
  - *then* a (*P*,*Q*,*R*) Tucker3 model fit the data appropriately



Unfold: *Wold, Geladi, Esbensen, Öhman. Principal component- and PLS-analyses generalized to multi-way (multi-order) data arrays. Copenhagen Symposium on Applied Statistics:249-277, 1986.*

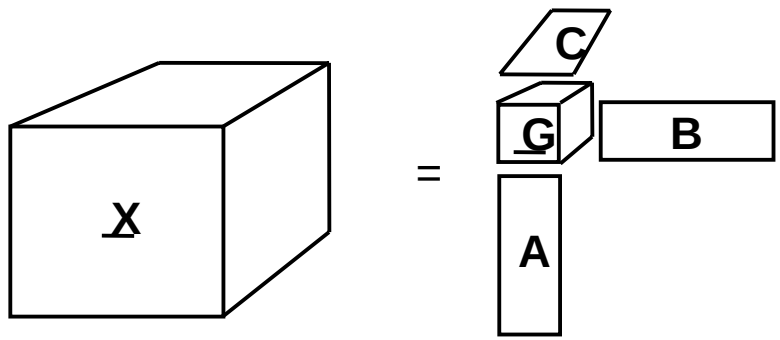
Tucker3 vs PARAFAC

- Differences from PARAFAC/PCA:
  - The number of components can vary in **A**, **B**, and **C**!
  - **G** is not superdiagonal
  - Tucker loadings not unique (only subspace) = rotational freedom
  - Tucker loadings orthogonal => variance-partitioning
  - PARAFAC best rank *F* model, but does not describe the part of data within that subspace!
  - Tucker best subspace *F* model but not rank *F* !!



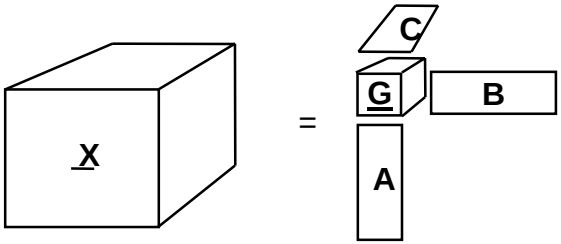
Tucker3 - algorithm

1. Initialize **B** and **C** (e.g. from SVD)
2. **A** equals first *P* left singular vectors of $\mathbf{X}^{(I\times JK)}(\mathbf{C}\otimes\mathbf{B})$
3. **B** equals first *Q* left singular vectors of $\mathbf{X}^{(J\times IK)}(\mathbf{C}\otimes\mathbf{A})$
4. **C** equals first *R* left singular vectors of $\mathbf{X}^{(K\times IJ)}(\mathbf{B}\otimes\mathbf{A})$
5. Go to step 2 until relative changes are small
6. $\mathbf{G} = \mathbf{A}'\mathbf{X}(\mathbf{C}\otimes\mathbf{B})$



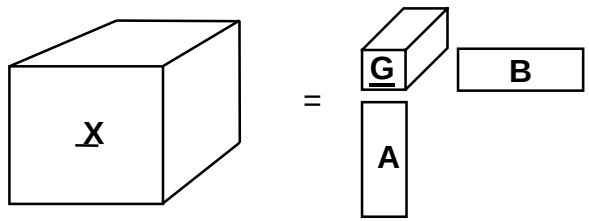
Other Tucker models

- Tucker3 has the number 3 because three modes are 'reduced'.
- Tucker2 and Tucker1 reduces two and one modes respectively



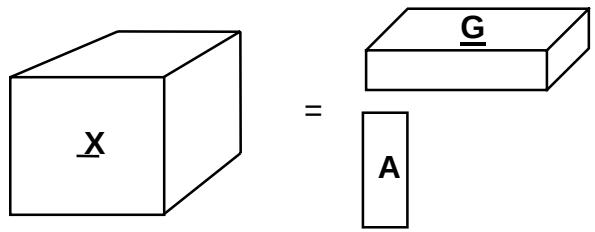
Tucker3

Tucker2 core often called Extended Core Array



Tucker2

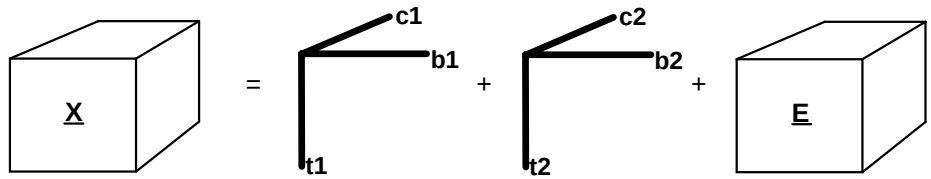
Tucker1 is identical to PCA on matricizes **X**



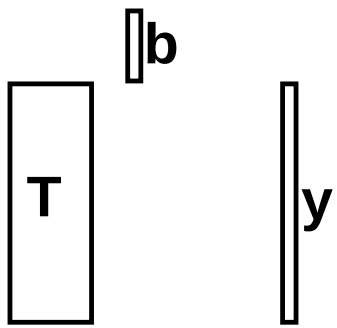
Tucker1

Multilinear PLS regression

- N-PLS



  - Use a trilinear (PARAFAC-like) model of **X** but such that the scores are predictive of **y**.



Bro. Multiway calibration. Multi-linear PLS. Journal of Chemometrics 10:47-61, 1996.

Multilinear PLS regression

- **Two-way PLS principle**
  - Find **w** (||**w**||=1) and **q** (||**q**||=1) such that the one-component models of **X** = **tw**' + $\mathbf{E}_X$ and **Y** = **uq**' + $\mathbf{E}_Y$ have maximal covariance (**t**'**u**)
  - Make regression model to predict **u** from **t**
  - Subtract the model from **X** and predictions from **Y**
  - Proceed with the next component from residuals

Multilinear PLS regression

- **Two-way PLS: how to do it - one y.**

Max covariance

$$\max_{\mathbf{w}} \left[\operatorname{cov}(\mathbf{t},\mathbf{y}) \;\middle|\; \mathbf{t} = \mathbf{X}\mathbf{w} \right] =$$

LS model

$$\max_{\mathbf{w}} \left[\mathbf{t}'\mathbf{y} \;\middle|\; \mathbf{t} = \mathbf{X}\mathbf{w} \right] =$$

$$\max_{\mathbf{w}} \left[\mathbf{y}'\mathbf{X}\mathbf{w} \right] =$$

$$\max_{\mathbf{w}} \left[\mathbf{z}'\mathbf{w} \right] \Rightarrow$$

$$\mathbf{w} = \frac{\mathbf{X}'\mathbf{y}}{\|\mathbf{X}'\mathbf{y}\|}$$

Multilinear PLS regression

Three-way PLS: how to do it - one y.

$$\max_{\mathbf{w}^J\mathbf{w}^K}\left[\operatorname{cov}(\mathbf{t},\mathbf{y}) \;\middle|\; t_i=\sum_{j=1}^{J}\sum_{k=1}^{K}x_{ijk}w_j^J w_k^K\right]=$$

$$\max_{\mathbf{w}^J\mathbf{w}^K}\left[\sum_{i=1}^{I}t_i y_i \;\middle|\; t_i=\sum_{j=1}^{J}\sum_{k=1}^{K}x_{ijk}w_j^J w_k^K\right]=$$

$$\max_{\mathbf{w}^J\mathbf{w}^K}\left[\sum_{i=1}^{I}\sum_{j=1}^{J}\sum_{k=1}^{K}y_i x_{ijk}w_j^J w_k^K\right]=$$

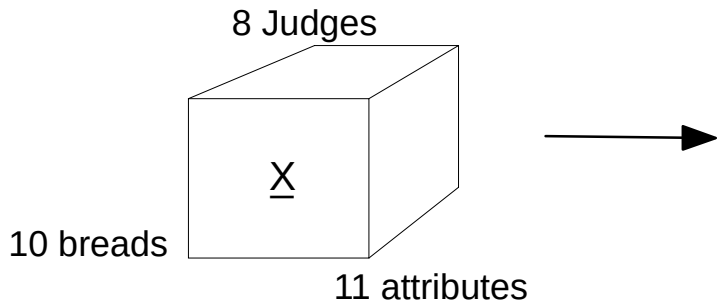
$$\max_{\mathbf{w}^J\mathbf{w}^K}\left[\sum_{j=1}^{J}\sum_{k=1}^{K}z_{jk}w_j^J w_k^K\right]=$$

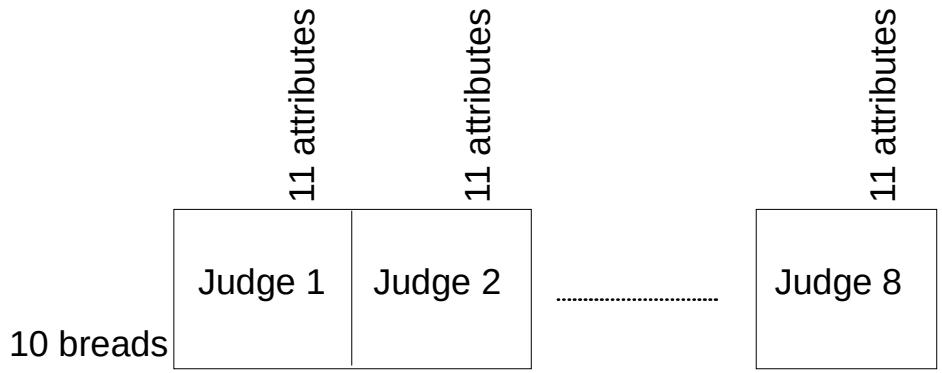
$$\max_{\mathbf{w}^J,\mathbf{w}^K}\left[(\mathbf{w}^J)'\mathbf{Z}\mathbf{w}^K\right]\Rightarrow$$

SVD on $\mathbf{Z}$

Example sensory data

- Three-way, two-way:
- Does it make a difference?
  - 5 breads (in replicates) × 11 attributes × 8 judges
  - Data due to Magni Martens

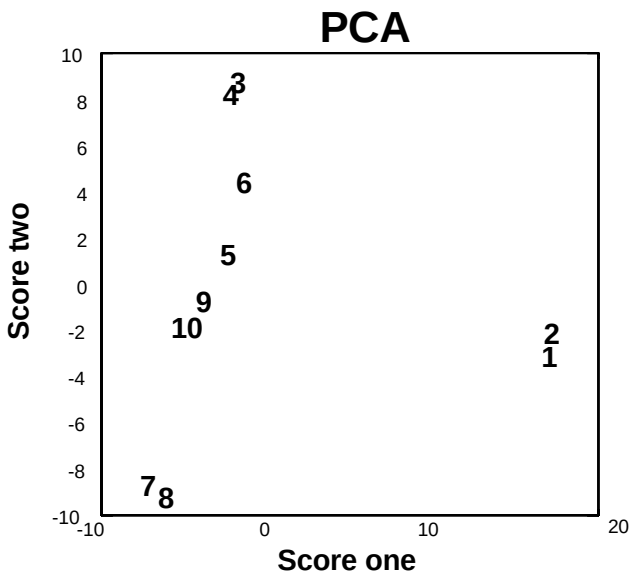


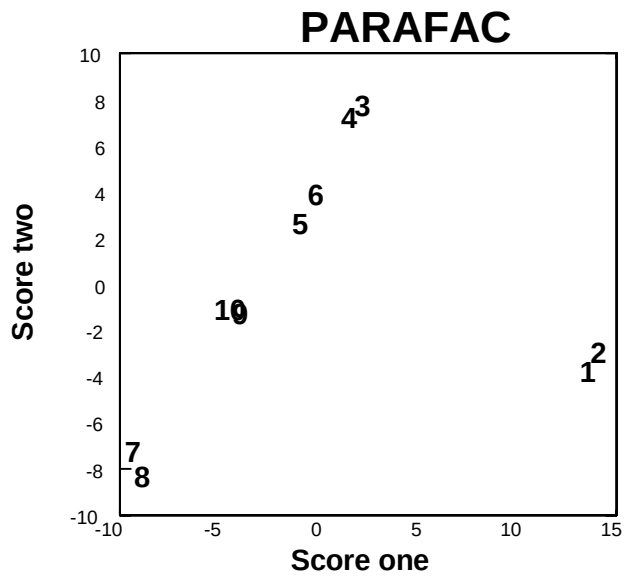


Sensory data

- Scores from bilinear PCA and trilinear PARAFAC

Three-way more robust because of 'stronger' structural model

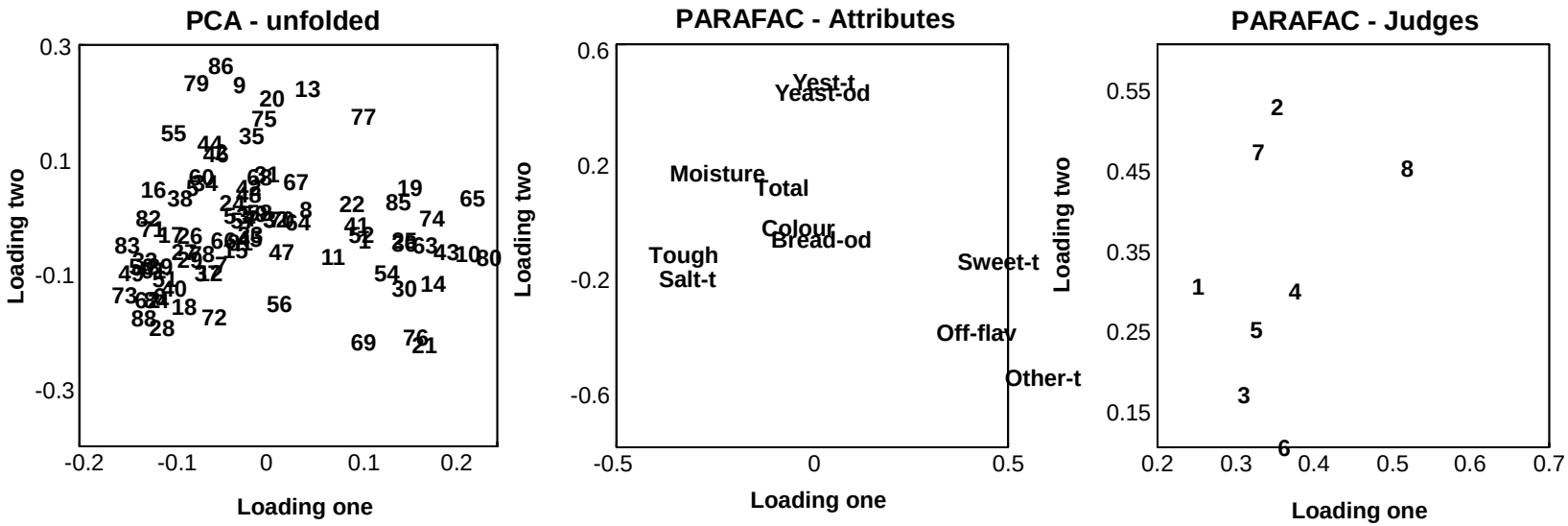




- Similar but note that replicates are closer for PARAFAC

Sensory data

- Loadings from bilinear PCA and trilinear PARAFAC



PARAFAC 19 loading-elements per component

PCA 88 loading-elements per component!

Sensory data

- Calibration - predict salt content
  - 25% improvement!!

Three-way more predictive because less overfit

	LV	Variation explained /%				RMSE	
		X cal.	X val.	Y cal.	Y val.	Y cal.	Y val.
unfold-PLS	1	43	25	80	62	0.21	0.29
	2	61	38	95	76	0.10	0.23
	3	74	49	100	84	0.03	0.19
	4	78	49	100	84	0.01	0.19
	5	84	50	100	84	0.00	0.19
	6	87	52	100	84	0.00	0.19
Trilinear PLS	1	31	22	75	60	0.23	0.30
	2	46	36	93	82	0.12	0.20
	3	54	44	98	91	0.07	0.15
	4	57	46	100	91	0.03	0.14
	5	60	47	100	90	0.02	0.15
	6	61	47	100	90	0.00	0.15

Second order calibration

- Two-way classical least squares mixture model **X** = **CS**',
  - E.g.
  - **X** the measured spectra, **C** the true concentrations, **S** the pure spectra
- If pure spectra, **S**, known, **C** determined by a simple least squares regression step – $\mathbf{C} = \mathbf{X}(\mathbf{S}')^{+}$
- Thus, if all spectra known quantitation possible and no calibration samples are needed

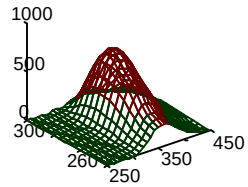
How to do it

- In three-way PARAFAC the model is $\mathbf{X}_k = \mathbf{A}\mathbf{D}_k\mathbf{B}'$,
  - $\mathbf{X}_k$ the measured spectra at occasion *k*
  - **A** the true concentrations
  - **B** the pure spectra
  - **C** (rows from $\mathbf{D}_k$) pure spectra/profiles etc.
- Pure spectra not necessary. They are found by decomposing with PARAFAC
- Thus, without prior knowledge quantitation possible and no calibration samples are needed except for fixing the scale

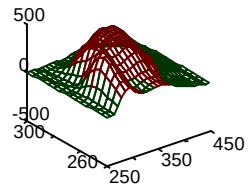
Fluorescence example

- **Calibration set**: *One* sample with *one* analyte (2.67 µM Trp)
- **Test set**: Two samples with three analytes each (Trp, Tyr, Phe)
- Three-component PARAFAC model fitted to the fluorescence data (3×201×61):
  - **A** - estimated concentrations
  - **B** - estimated emission spectra
  - **C** - estimated excitation spectra

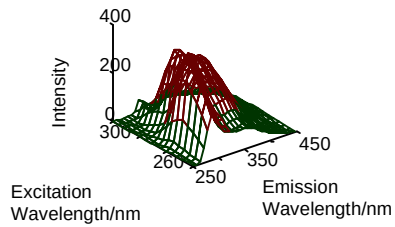
Standard (Trp)

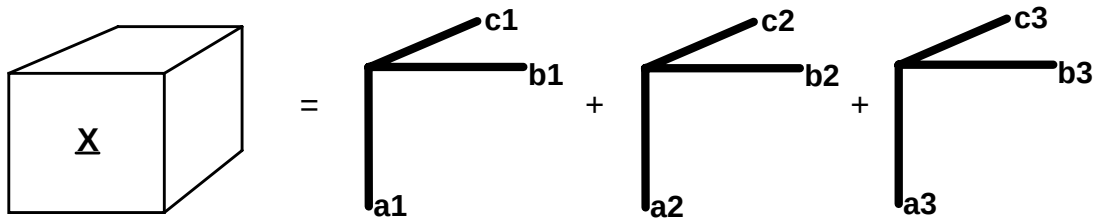


Unknown 1 (Tyr,Trp,Phe)



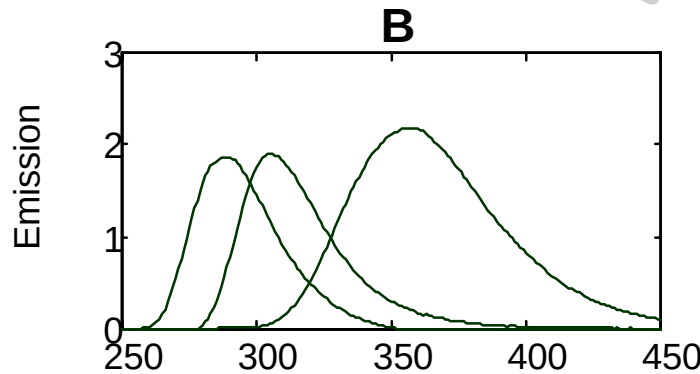
Unknown 2 (Tyr,Trp,Phe)

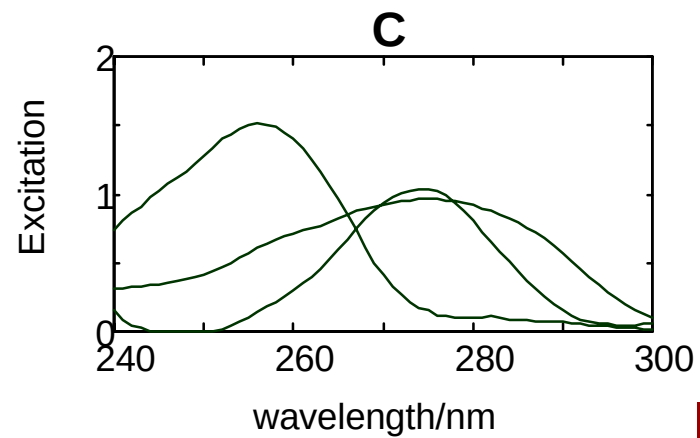




Fluorescence example

- Spectral loadings resemble pure spectra
- Sample scores - estimated concentration





$$\mathbf{A} = \begin{array}{c} \begin{matrix} \textbf{Trp} & \textbf{Tyr} & \textbf{Phe} \end{matrix} \\ \begin{bmatrix} 2.67 & 0 & 0 \\ 1.52 & 1.30 & 1.29 \\ .86 & 1.12 & 1.06 \end{bmatrix} \end{array}, \quad \mathbf{reference} = \begin{bmatrix} 2.67 \\ 1.58 \\ .88 \end{bmatrix}$$

Prediction

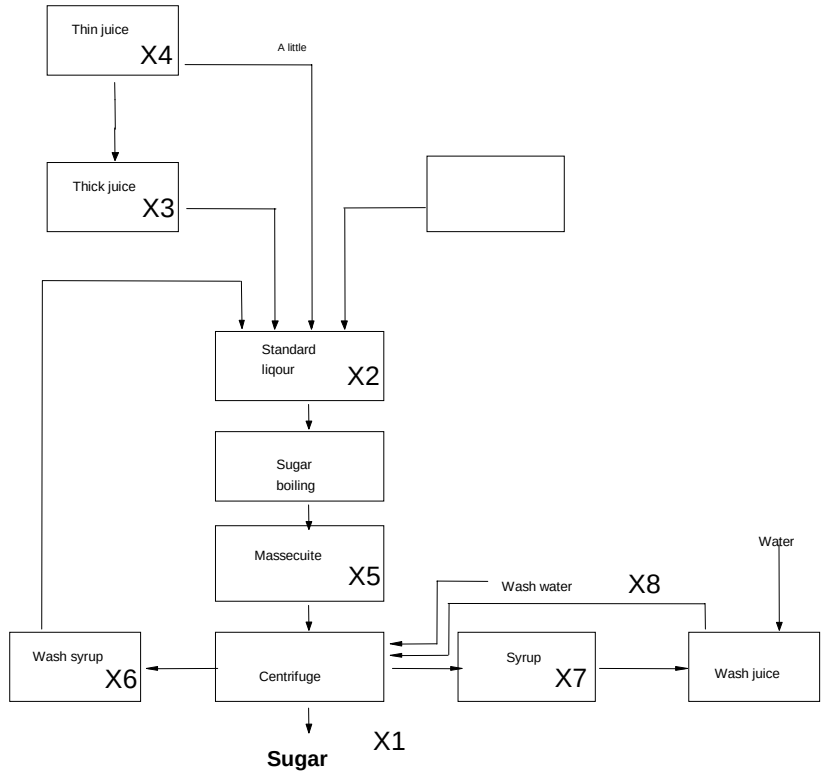
- **How to make a prediction model**
  - Using sub-space regression, e.g. N-PLS
  - Using ‘second-order calibration’ mostly with PARAFAC

- **Significant difference**
  - Predictions based on ordinary *regression* models only work if all interferents are varying independently in the calibration data ⇒
    - Many samples needed
    - Only similar samples can be predicted
  - PARAFAC models work with only *one* calibration sample and with *unknown uncalibrated* interferents

Sugar processing

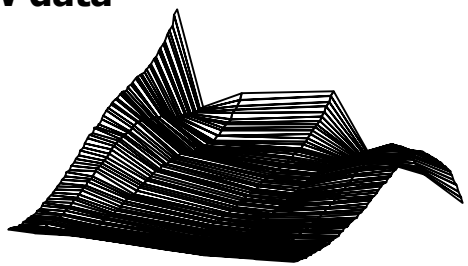
Sugar made from beets

- End product sampled every 8'th hour during the three months of operation
- Measured spectrofluorometrically
- 260 samples



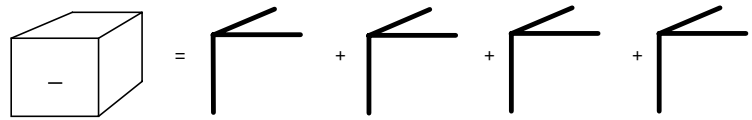
Sugar data model

Raw data

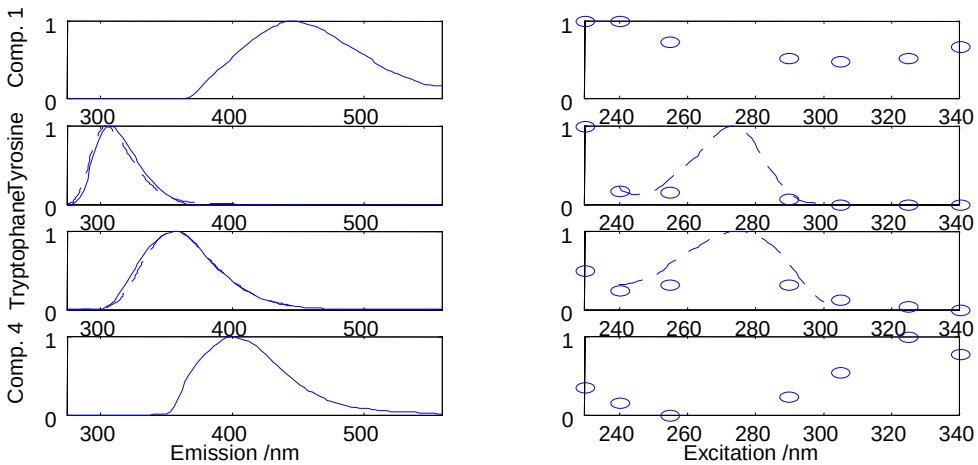


Raw data: 268 samples from a sugar campaign - fluorescence landscapes

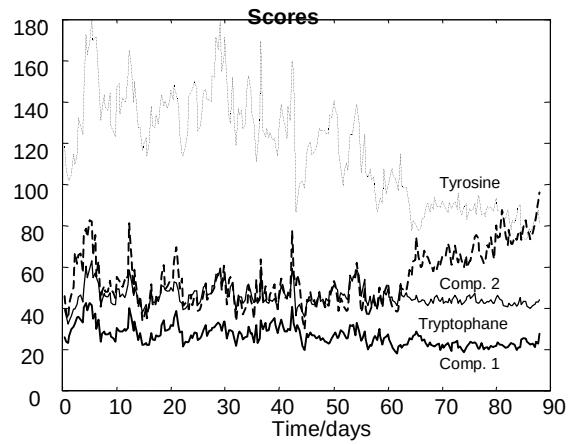
PARAFAC model



Deconvoluted spectra



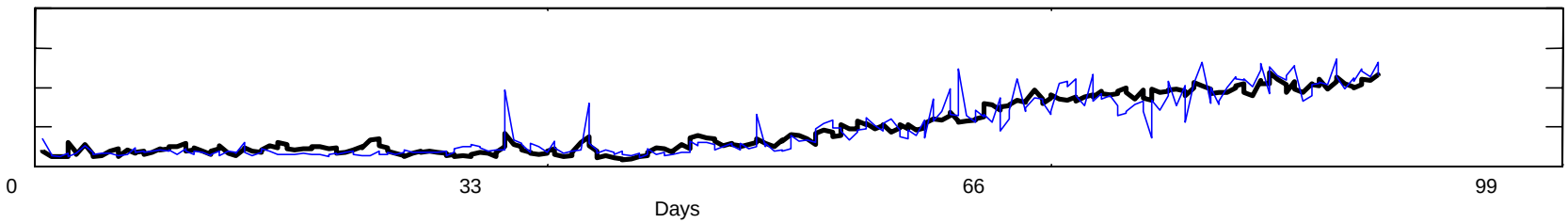
Concentrations



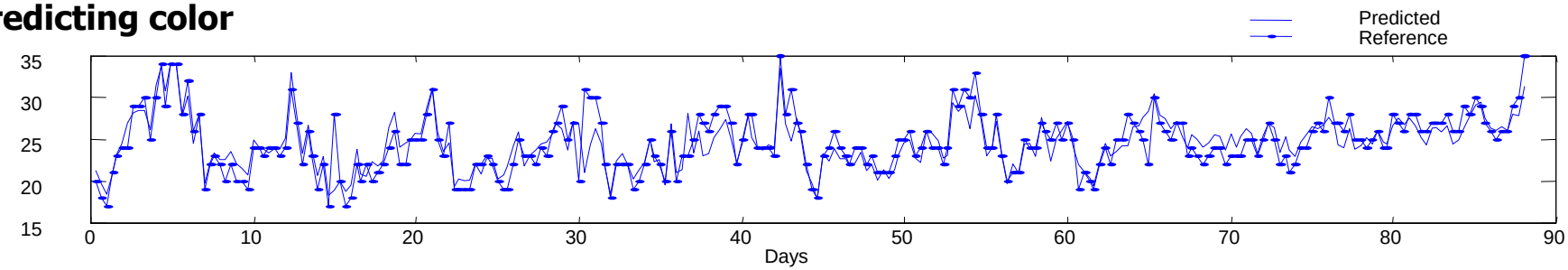
Using the model

- PARAFAC and fluorescence: unique combination of *multivariate process control* and *process analytical chemistry*
- The process can be monitored and controlled on a *chemical* level

Predicting CaO



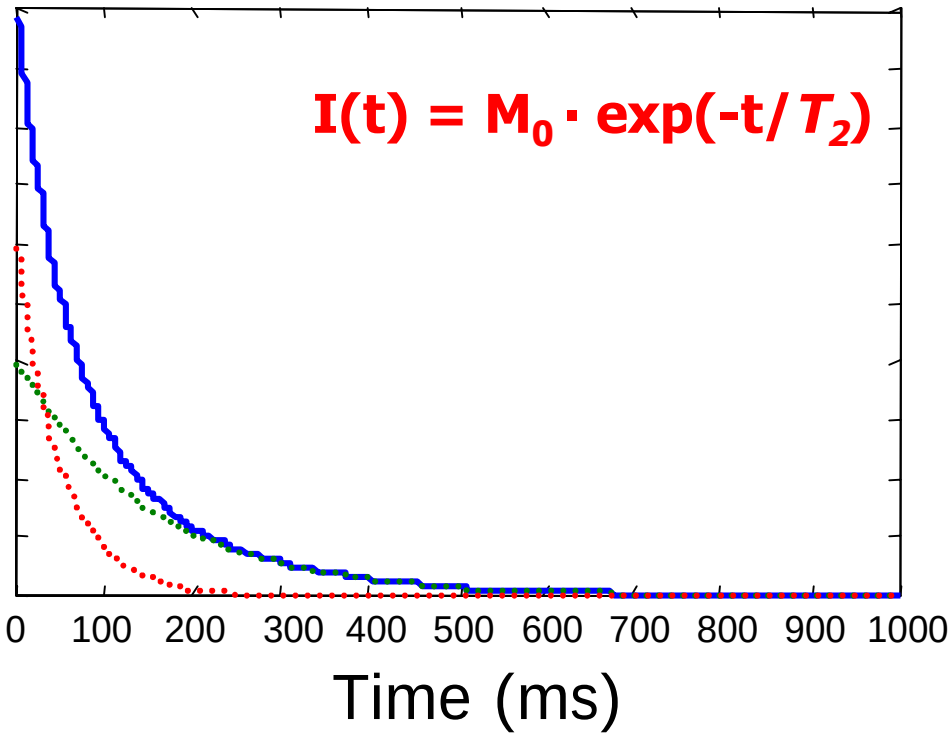
Predicting color



Resolving two-way exponentials

- **Two-way data but with very special structure in loadings**

$$\mathbf{X} = \mathbf{AB}' = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 27 & 9 & 3 & 1 \\ 16 & 8 & 4 & 2 \end{bmatrix}$$



Sample 1	Sample 2
27	16
9	8
3	4
1	2

SLICING - Pseudo Three-way Data

$$X_1 = AD_1B' = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\begin{bmatrix} 3 & 0 \\ 0 & 2 \end{bmatrix}\begin{bmatrix} 9 & 3 & 1 \\ 8 & 4 & 2 \end{bmatrix}$$

SLAB 1 (X_1)

Sample 1	Sample 2
27	16
9	8
3	4

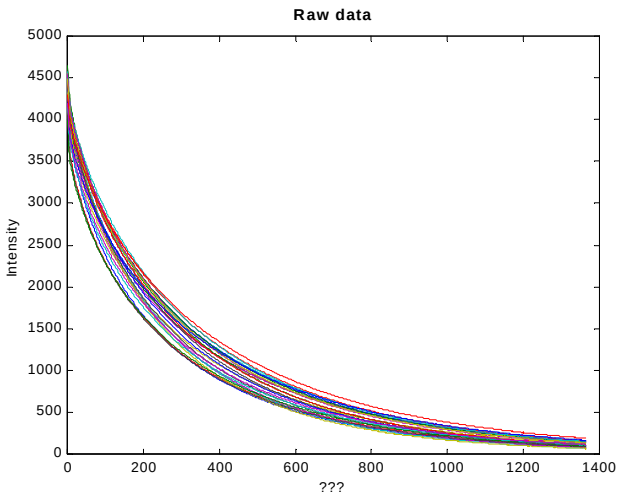
$$X_2 = AD_2B' = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}\begin{bmatrix} 9 & 3 & 1 \\ 8 & 4 & 2 \end{bmatrix}$$

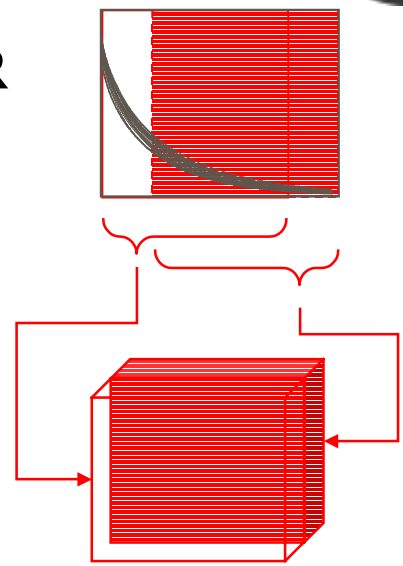
SLAB 2 (X_2)

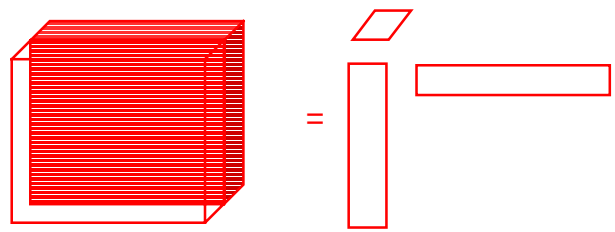
Sample 1	Sample 2
9	8
3	4
1	2

Example: Sensory quality

- **Predicting cooked potato quality from NMR on raw data**
  - Potatoes cooked and served hot. Ten assessors evaluate texture profile, e.g. mealiness
  - *Raw* (!) potato measured by NMR (CPMG pulse sequence)



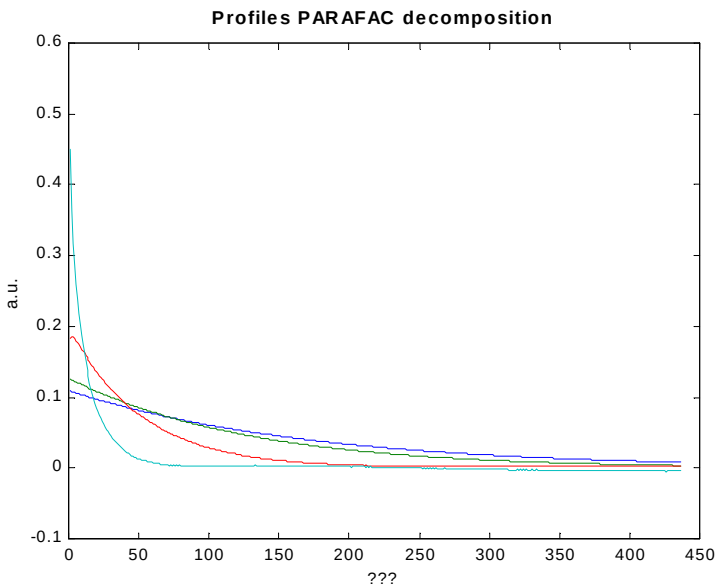


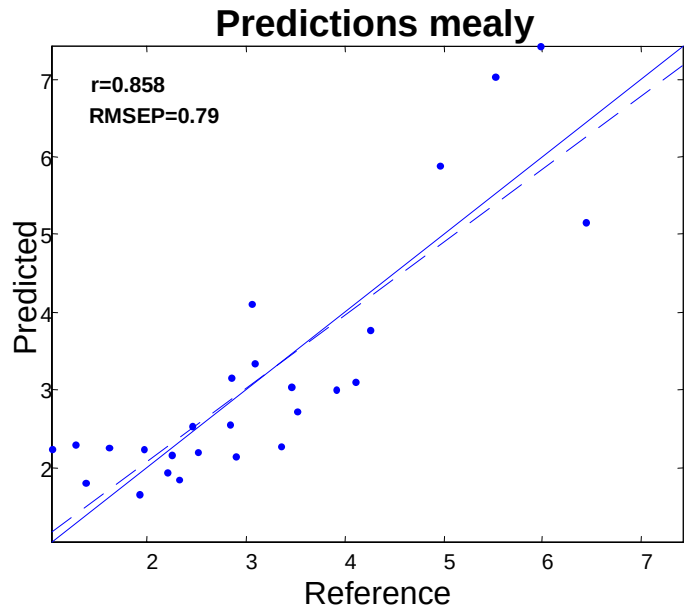


A. K. Thybo et. al., *Food Science and Technology,* 33 (2):103-111, 2000.

Example: Sensory quality

- **Decomposing NMR into meaningful latent variables**
  - PARAFAC on lagged data enables meaningful decomposition
  - Four components adequate for describing NMR data
- **Predictions of mealiness from NMR**
  - Predicting sensory quality of *cooked* potatoes from amount of latent variables (i.e. directly from NMR of *raw* potatoes)





Using constraints

- **Using constraints in multi-way modeling**

- **Example**
  - Instead of 'PCA': $\| \mathbf{X} - \mathbf{AB}' \|$
  - fit the model: $\| \mathbf{X} - \mathbf{AB}' \|$,

    subject to **A** and **B** are nonnegative

- Constraints are essential in two-way curve resolution because the model is unidentified

- In three-way curve resolution the model is often unique but constraints are still useful

Why constraints?

- **Obtain sensible parameters**
  - Ex.: Require chromatographic profiles to have but one peak
- **Obtain unique solution**
  - Ex.: Use selective channels in data to obtain uniqueness
- **Test hypotheses**
  - Ex.: Investigate if tryptophane is present in sample
- **Avoiding degeneracy and numerical problems**
  - Ex.: Enabling a PARAFAC model of data otherwise inappropriate for the mode
- **Speed up algorithms**
  - Ex.: Use truncated bases to reexpress problem by a smaller problem
- **Enable quantitative analysis of qualitative data**
  - Ex.: Incorporate sex and job type in a model for predicting income

Typical constraints

	Spectroscopy	Chromatography	FIA	Kinetics
Auto- & cross correlation	✔	✔	✔	✔
Uncertainty	✔	✔	✔	✔
Nonnegativity	✔	✔	✔	✔
Unimodality	(✔)	✔	✔	
Selectivity		✔	✔	
Smoothness	✔	✔	✔	✔
Known spectra	✔			
...				

FIA example

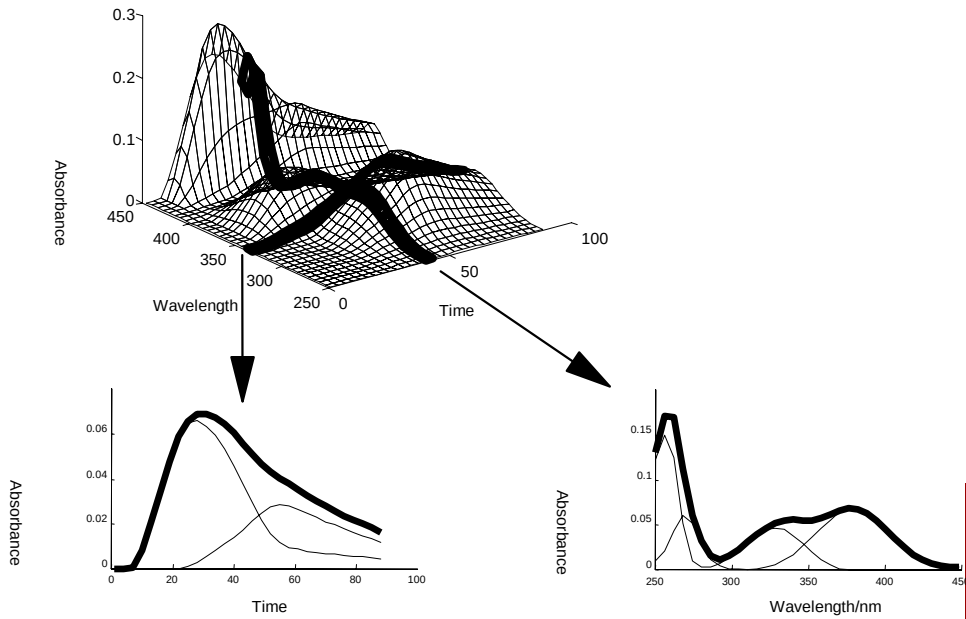
Model of a single sample with one analyte

- Samples of 2, 3, 4-HBA (hydroxy benzaldehyde) detected by UV-VIS in FIA system with pH-gradient imposed
- Every spectrum is a sum of acidic and basic spectrum. Same holds for time profiles. Only sums are measured
- Model not important here.
  PARALIND : $\mathbf{X}_k = \mathbf{A}\mathbf{H}\mathbf{D}_k\mathbf{B}'$

Data

Concentrations

Pure spectra

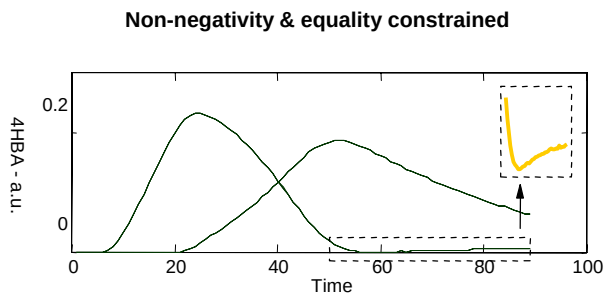


Effect of constraints

- Eq : Equality of summed profiles
- NNLS : Non-negativity of all parameters
- ULSR : Unimodality of FIAgrams/time profiles
- Fix : Fixing purely acidic/basic times to only reflect acidic/basic analytes

SPECTRA	2HBA acidic	2HBA basic	3HBA acidic	3HBA basic	4HBA acidic	4HBA basic
Eq	0.9893*	0.9871*	0.9689*	0.7647*	0.9106*	0.9211*
NNLS	0.9944	0.9117*	0.9952	0.9241	0.9974	0.9977
NNLS/Eq	0.9946	0.9312*	0.9953	0.9988	0.9965	0.9971
NNLS/ULSR/Eq	0.9946	0.9590*	0.9953	0.9989	0.9966	0.9943
NNLS/ULSR/Fix/Eq	0.9946	0.9989	0.9954	0.9986	0.9961	0.9977

Correlations



Problems to deal with

- **Algorithms**
  - Tucker, N-PLS are unproblematic, even for large data sets
  - PARAFAC problematic
    - Multiple local minima
    - 2-factor degeneracy (model-problem): no solution?
    - Solution sensitive to correct dimensionality (not sequential)
    - Slow convergence

- **Other problems currently considered**
  - Statistical measures (rank/DoF problems)
  - Diagnostics for choosing the number of components
  - Handling highly structured error covariances
  - Handling huge amounts of missing data

Concluding remarks

- **Uniqueness** (mainly PARAFAC)
  - Pure spectra
  - Second order advantage
    - No interferents, only one standard
- **Better structural model**
  - Robustness/noise reduction
  - Simpler model
  - Interpretation
  - Better predictions

Concluding remarks

References

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- Leurgans, Ross. Statist. Sci., 1992, **7**, 289
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- Bro. J. Chemom. 1996, **10**, 47
- Bro, Chemom.Intell.Lab.Syst., 1997, **38**, 149
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Software & info

- http://www.models.kvl.dk (Free matlab code'n'course, database of papers)
- http://www.ece.umn.edu/users/nikos/public_html/3SPICE/3SPICEmain.html
  TRIPLE SPICE – Multi-way analysis in signal processing – matlab, presentations etc.
- http://www.eigenvector.com (Matlab code)
- http://www.fsw.leidenuniv.nl/~kroonenb (Stand alone)