Can one recover the underlying spectral data matrix from a given Borgen plot?

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Abstract

In Multivariate Curve Resolution (MCR), Borgen plots represent the regions of feasible pure component profiles underlying spectral mixture data. A Borgen plot can be constructed geometrically in the low-dimensional U- and V-spaces if the so-called outer polygon (representing nonnegativity constraints) and the inner polygon (that is the convex hull of the data representing points) are given.

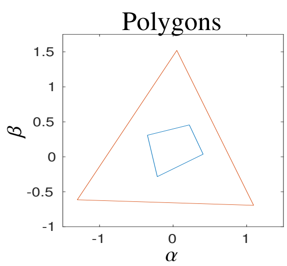
This paper asks whether it is possible to construct spectral data from the data representing points spanning the polygons, and thus reconstruct the data from the associated Borgen plot. A partially positive answer is given.

Keywords: multivariate curve resolution, Borgen plots

1. Introduction

In 1985 Borgen and Kowalski [3] introduced the geometric construction of Borgen plots for the analysis of the factor ambiguity problem in multivariate curve resolution. Starting from a given spectral data matrix D, Borgen plots can geometrically be constructed within the low-dimensional abstract spaces spanned by the left and right singular vectors of D. These spaces are often called the U- and the V-space. In a first step, the inner and outer polygons, which are denoted INNPOL and FIRPOL in [3], are determined in the U- and the V-space. The vertices of the inner polygons represent certain projections of the rows or columns of D in the spaces spanned by the singular vectors, and the outer polygons represent the nonnegativity constraints in the respective spaces. The feasible regions can then be constructed by the so-called triangle rotation algorithm. These regions are the sets of the vertices of all triangles that enclose the inner polygon and that are enclosed in the outer polygon. For an in-depth introduction to Borgen plots and their construction we refer to [1, 3, 7, 11, 12, 15].

Here, we consider the question whether there is a way back from the representation in the abstract plane to an underlying spectral data matrix. To explain this question, we have reproduced the two polygons as used by Borgen and Kowalski in their exclusive example, see Fig. 6 in [3]. The entries of the spectral data matrix determine the vertices of the inner polygon (here a quadrangle) and the edges of the outer polygon (here a triangle). However, Borgen and Kowalski do not present the matrix in [3]. Is it nevertheless possible to find a matrix that leads to these polygons? If such a matrix exists, we call it a *generator matrix*. Fig. 1 shows the abstract plane together with a 4×3 generator matrix with smallest possible dimensions as found by our algorithms.



Generator matrix

$$\longrightarrow \quad D = \begin{pmatrix} 0.682 & 0.293 & 1 \\ 0.142 & 0.0374 & 0.086 \\ 0.739 & 0.669 & 0.229 \\ 0.328 & 0.889 & 0.629 \end{pmatrix}$$

Figure 1: The classical polygons INNPOL (blue) and FIRPOL (red) as used by Borgen and Kowalski and a possible generator matrix of smallest dimensions as found by our algorithm. It is a 4×3 matrix according to the 4 vertices of the inner polygon and the 3 edges of the outer polygon.

The inner and outer polygons can be constructed not only in the 2D plane of a Borgen plot. In general, for chemical systems with s chemical species the so-called Area of Feasible Solutions (AFS) is constructed in the $(s-1)$-dimensional spaces of the left and right singular vectors with a fixed contribution equal to 1 from the first right (or left) singular vector. Considering such a fixed contribution from the first singular vectors justifies working in $(s-1)$-dimensional spaces instead of s-dimensional spaces. For $s > 3$ the polygons become inner and outer polytopes, see

[12, 15] for details. The following analysis is formulated for arbitrary $s \geq 3$ and also applies to the elementary case of Lawton-Sylvestre plots with $s = 2$.

1.1. Polygons and polytopes in the U- and V-space

Let a nonnegative (spectral) data matrix $D \in \mathbb{R}^{k \times n}$ be given, which has a nonnegative factorization $D = CS^T$ with the concentration factor $C \in \mathbb{R}^{k \times s}$ and the spectral factor $S \in \mathbb{R}^{n \times s}$. The ranks of these three matrices are equal to s. The truncated SVD is $D = U\Sigma V^T$ with the orthogonal matrices of left and right singular vectors $U \in \mathbb{R}^{k \times s}$ and $V \in \mathbb{R}^{n \times s}$. The $s \times s$ diagonal matrix Σ has the singular values σ_i on its diagonal. It is a known fact that then there exists a regular $s \times s$ matrix T that allows to express the pure component factors as $C = U\Sigma T^{-1}$ and $S^T = TV^T$. See [1, 3, 7, 11] for details.

These matrices determine the outer polytope $\mathcal{F}_S$ and the inner polytope $\mathcal{I}_S$ in the V-space. For $s = 3$ the polytopes are 2D polygons and for $s = 4$ the geometric objects are 3D polyhedra

$$
\begin{aligned}
\mathcal{I}_S &= \text{convhull}(\{a_i \in \mathbb{R}^{s-1}, i = 1, \ldots, k\}), \quad \text{with} \quad a_i = \frac{((U\Sigma)(i, 2:s))^T}{(U\Sigma)(i, 1)} \\
\mathcal{F}_S &= \{x \in \mathbb{R}^{s-1}, \text{ so that } (1, x^T)V^T \geq 0\}.
\end{aligned} \tag{1}
$$

A similar construction holds in the U-space. By duality [5, 9, 12] the vertices of the inner polytope $\mathcal{I}_C$ are dual to the edges of $\mathcal{F}_S$. Analogously the nonnegativity constraints on C lead to the outer polytope $\mathcal{F}_C$ which is dual to $\mathcal{I}_S$

$$
\begin{aligned}
\mathcal{I}_C &= \text{convhull}(\{b_j \in \mathbb{R}^{s-1}, j = 1, \ldots, n\}), \quad \text{with} \quad b_j = \frac{(V(j, 2:s))^T}{V(j, 1)} \\
\mathcal{F}_C &= \{y \in \mathbb{R}^{s-1}, \text{ so that } U\Sigma \begin{pmatrix} 1 \\ y \end{pmatrix} \geq 0\}.
\end{aligned} \tag{2}
$$

This representation in the U- and V-space is called the *low-dimensional representation*. The a_i and b_j are called *data representing points* or just *data points*, because they are related to the rows and columns of the data matrix D. The polytopes $\mathcal{I}_C$ and $\mathcal{I}_S$ are sufficient to determine all polytopes as $\mathcal{I}_C$ is dual to $\mathcal{F}_S$ and $\mathcal{I}_S$ is dual to $\mathcal{F}_C$.

1.2. Problem definition

Having defined the data points a_i and b_j and the polytopes $\mathcal{I}_S$, $\mathcal{F}_S$, $\mathcal{I}_C$ and $\mathcal{F}_C$, we can give the Borgen plot reconstruction problem a precise form.

Problem 1.1 (From a_i and b_j to D). *Let only the data points $a_i, b_j \in \mathbb{R}^{s-1}$ by Eq. (1) and Eq. (2) be given. We do not assume to know the original matrix D that defines these data points. Is it possible to find a rank-s generator matrix $\hat{D}$ so that its data points $\hat{a}_i$ and $\hat{b}_j$ have the same convex hull than the original data points? Hence D and $\hat{D}$ have the same four polytopes (the outer polytopes by duality) and $\hat{D}$ reproduces the original Borgen plot/AFS.*

Why do we ask this question? First, we want to know if the U- and V-space representation of the data points inevitably leads to a loss of information contained in the spectral data matrix. Second, in theoretical studies, we are sometimes faced with the problem of whether certain polytopes $\mathcal{I}_S$, $\mathcal{F}_S$, $\mathcal{I}_C$ and $\mathcal{F}_C$ or a resulting AFS of a certain shape (e.g., hand-drawn polytopes/AFS in 2D or 3D) can be meaningful. In other words, does a corresponding generator matrix exist? This brings us to a second problem.

Problem 1.2 (From hand-drawn polytopes to D). *Let one polytope (the outer polytope) enclose a second polytope (the inner polytope) in the $\mathbb{R}^{s-1}$. The inner polytope contains the origin (the null vector). The polytopes are only given in terms of their shape, e.g., by the Euclidean coordinates of their vertices, and we do not suppose that the polytopes originate from a certain data set. Is it possible to find a corresponding rank-s generator matrix D that reproduces these polytopes?*

These two problems make different assumptions about what is given. Problem 1.1 assumes that all data points are known, while in Problem 1.2 we do not necessarily know all data points, but at least those that span the polytopes.

1.3. Organization of this paper

First, Problem 1.1 is traced back to a nonlinear system of equations with additional nonnegativity constraints, see Sec. 2. Solutions can be found by numerical optimization. Second, the reconstruction problem of polytopes with $0 \in \mathcal{I} \subset \mathcal{F}$ without assuming that they originate from a matrix D is posed in Problem 1.2. A solution is suggested in Sec. 3. In both cases, the goal is to construct a generator matrix. This paper presents an analytical solution approach together with numerical solution algorithms.

2. Solution of problem 1.1 by constructing a generator matrix

Our starting point is the data point representation

$$
\begin{aligned}
a_i &= \frac{((U\Sigma)(i,2:s))^T}{(U\Sigma)(i,1)} = \frac{((DV)(i,2:s))^T}{(DV)(i,1)} \in \mathbb{R}^{s-1}, \qquad i=1,\dots,k,\\
b_j &= \frac{(V(j,2:s))^T}{V(j,1)} = \frac{((D^T U\Sigma^{-1})(j,2:s))^T}{V(j,1)} \in \mathbb{R}^{s-1}, \qquad j=1,\dots,n.
\end{aligned} \tag{3}
$$

We assume that we know all data points a_i and b_j, but that we do not know the SVD factors U, Σ and V. We study the systems of equations (3) that relate the factors of the SVD to the data point representations given by a Borgen plot together with the orthogonality constraints on the columns of U and V. Each solution of this system of equations allows us to obtain a possible generator matrix $\hat{D} = U\Sigma V^T \in \mathbb{R}^{k\times n}$ that generates the data points. The a_i are the representations of the rows of D in terms of the scaled right singular vectors, and the b_j are the representations of the columns of D in terms of the scaled left singular vectors. These representations are scaled in a way that the contributions by the first left/right singular vector are equal to 1 and all these first components are truncated by accessing only to the components $2:s$ (this is expressed by using the colon notation). Such a scaling is possible because the Perron-Frobenius spectral theory of nonnegative irreducible matrices guarantees a nonzero contribution to any nonnegative profile from the first left/right singular vectors [8, 13]. Hence the data points a_i and b_j are $(s-1)$-dimensional vectors, namely projections from the $\mathbb{R}^s$. However, the scaling removes the information contained in the first left singular vector u_1 and the first right singular vector v_1. Can this information be recovered? This problem is discussed next.

Assuming that the data points $b_j = (b_{j1},\dots,b_{j(s-1)})^T$ are given, we can write the matrix V of the right singular vectors as follows

$$
V = (\mathrm{v}_1,\dots,\mathrm{v}_s) = \begin{pmatrix} \mathrm{v}_{11} & \mathrm{v}_{11}\, b_1^T \\ \vdots & \vdots \\ \mathrm{v}_{n1} & \mathrm{v}_{n1}\, b_n^T \end{pmatrix} = \begin{pmatrix} \mathrm{v}_{11} & \mathrm{v}_{11}\cdot b_{11} & \dots & \mathrm{v}_{11}\cdot b_{1,s-1} \\ \vdots & \vdots & & \vdots \\ \mathrm{v}_{n1} & \mathrm{v}_{n1}\cdot b_{n1} & \dots & \mathrm{v}_{n1}\cdot b_{n,s-1} \end{pmatrix} \in \mathbb{R}^{n\times s}. \tag{4}
$$

The n components of the first right singular vector v_1 are the unknowns. The orthogonality of V leads to the equations

$$
\begin{aligned}
(\mathrm{v}_1,\mathrm{v}_1) &= \sum_{j=1}^{n} \mathrm{v}_{j1}^2 = 1, \\
(\mathrm{v}_1,\mathrm{v}_\ell) &= \sum_{j=1}^{n} \mathrm{v}_{j1}^2 b_{j,\ell-1} = 0, \qquad && \ell = 2,\dots,s, \\
(\mathrm{v}_r,\mathrm{v}_t) &= \sum_{j=1}^{n} \mathrm{v}_{j1}^2 b_{j,r-1} b_{j,t-1} = \delta_{rt}, \qquad && r,t = 2,\dots,s,
\end{aligned} \tag{5}
$$

where $(\cdot,\cdot)$ is the Euclidean inner product. Therein δ_{rt} is the Kronecker delta that equals 1 if $r=t$ and 0 otherwise. The number of equations is $1+(s-1)+(s-1)s/2 = (s^2+s)/2$.

Second, the left singular vectors can be expressed in terms of the data points $a_i = (a_{i1},\dots,a_{i(s-1)})^T$ as follows

$$
U = (U\Sigma)\Sigma^{-1} = \sigma_1 \begin{pmatrix} u_{11} & u_{11} a_1^T \\ \vdots & \vdots \\ u_{k1} & u_{k1} a_k^T \end{pmatrix} \Sigma^{-1} = \sigma_1 \cdot \begin{pmatrix} u_{11} & u_{11}\cdot a_{11} & \dots & u_{11}\cdot a_{1,s-1} \\ \vdots & \vdots & & \vdots \\ u_{k1} & u_{k1}\cdot a_{k1} & \dots & u_{k1}\cdot a_{k,s-1} \end{pmatrix} \Sigma^{-1} \in \mathbb{R}^{k\times s} \tag{6}
$$

where $\Sigma = \operatorname{diag}(\sigma_1,\dots,\sigma_s)$. The k unknown components of the first left singular vector u_1 satisfy the orthogonality conditions on U, namely

$$
\begin{aligned}
(u_1,u_1) &= \sum_{i=1}^{k} u_{i1}^2 = 1, \\
(u_1,u_\ell) &= \sum_{i=1}^{k} u_{i1}^2 a_{i,\ell-1} = 0, \qquad && \ell = 2,\dots,s, \\
(u_r,u_t) &= \frac{\sigma_1}{\sigma_r}\,\frac{\sigma_1}{\sigma_t} \sum_{i=1}^{k} u_{i1}^2 a_{i,r-1} a_{i,t-1} = \delta_{rt}, \qquad && r,t = 2,\dots,s.
\end{aligned} \tag{7}
$$

Again, these are $(s^2 + s)/2$ equations so that the total number of equations is $s^2 + s$. In total, $s^2 + s$ equations are available to determine the $k+n$ components of $u_1 \in \mathbb{R}^k$, $\mathrm{v}_1 \in \mathbb{R}^n$ and the $s-1$ unknown ratios σ_1/σ_r for $r = 2, \ldots, s$. In most cases, the number of unknowns is much larger than the number of equations. Nonlinear numerical optimization can be used to find feasible solutions. This optimization has to take into account the constraint of componentwise positive vectors $u_1 > 0$ and $\mathrm{v}_1 > 0$, as required by the Perron-Frobenius theory for the so-called Perron vectors and that the singular values form a decreasing sequence in i. For such nonlinear, constrained optimization problems multiple solutions for the SVD factors can be expected. Therefore, multiple feasible solutions $\hat{D} = U\Sigma V^T$ are possible as generator matrices.

2.1. *Reconstruction of a generator matrix with smallest dimension for $s = 2$*

There is only one case with more equations than unknowns. This is a rank-2 matrix ($s = 2$) with the smallest matrix dimensions $k = n = 2$. Then the number of equations is 6 and the number of unknowns is $k + n + 1 = 5$. Then the original data matrix can be recovered up to scaling. The remaining scaling nonuniqueness of D does not affect the data reconstruction problem as the data vectors are not changed if D is substituted by a positive multiple of D; for a detailed explanation see Lemma Appendix A.4. In the case $s = 2$ the counterpart of a Borgen plot is the Lawton-Sylvestre plot [6]. In this simple case the inner and outer polytopes are just intervals that are each determined by their two endpoints. These endpoints are the scalar data points $a_i, b_j \in \mathbb{R}^1$ with $s - 1 = 1$. These points are shown in Fig. 2 for the V-space with $\mathcal{I}_S = [a_1, a_2]$ and $\mathcal{F}_S = [\tilde{b}_1, \tilde{b}_2]$. Using duality, the data points of $\mathcal{I}_C = [b_1, b_2]$ satisfy $b_i = -1/\tilde{b}_i$, $i = 1, 2$.

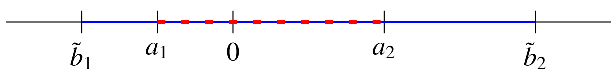


Figure 2: Lawton-Sylvestre plot in the V-space of the 2×2 matrix D. $\mathcal{I}_S$ is indicated by the dashed red line and $\mathcal{F}_S$ by the blue line.

Eq. (5) determines the two components of the first right singular vector v_1, and Eq. (7) determines the first left singular vector u_1. Their components are given by

$$\mathrm{v}_{11} = \left(1 - \frac{\tilde{b}_2}{\tilde{b}_1}\right)^{-\frac{1}{2}}, \qquad \mathrm{v}_{21} = \left(1 - \frac{\tilde{b}_1}{\tilde{b}_2}\right)^{-\frac{1}{2}},$$
$$u_{11} = \left(1 - \frac{a_1}{a_2}\right)^{-\frac{1}{2}}, \qquad u_{21} = \left(1 - \frac{a_2}{a_1}\right)^{-\frac{1}{2}}.$$

Thus D is uniquely determined, except for scaling, since only the ratio σ_1/σ_2 of the singular values is part of the system of equations (7). For this ratio we get

$$(\sigma_1/\sigma_2)^{-1} = \sqrt{-a_1 a_2}. \tag{8}$$

The square root can be taken since $\mathcal{I}_S = [a_1, a_2]$ with $a_1 < 0$ and $a_2 > 0$ (the null is in the interior of $\mathcal{I}$). The more general case $s = 2$ and $k, n > 2$ can be reduced to $k = n = 2$, since the endpoints of the intervals (the intervals are the inner polytopes) relate to only two (essential) data points. But then only a 2×2 generator matrix can be found.

2.2. *Solution of Problem 1.1 by numerical optimization*

Except for the trivial case $s = 2 = k = n$, the constrained nonlinear equations (5) and (7) are to be solved by numerical techniques. More precisely, Eq. (5) is a linear system in the squared variables v_{j1}^2, which is underdetermined if $(s^2 + s)/2 < n$, and Eq. (7) is a nonlinear system in the variables u_{i1} and the ratios σ_1/σ_r, $r = 2, \ldots, s$. A numerical solution can be found by numerical optimization under the constraints $u_1, \mathrm{v}_1 > 0$ and $\sigma_2/\sigma_1 \geq \ldots \geq \sigma_s/\sigma_1$. The goal is to find a generator matrix $\hat{D}$ that leads to the same data points a_i and b_j. Since σ_1 remains unknown, we scale the resulting matrix $\hat{D}$ so that its maximum entry is 1.

Fig. 3 shows a three-component model data set along with the data points in the U- and V-space marked by gray circles. The original data matrix D is a solution to the optimization problem, but there are also very different generator matrices, see Fig. 4. The results depend on the chosen optimization algorithm and the initial values for the first singular vectors and the first singular value. If we take the all-ones vectors $(1, \ldots, 1)^T$ as initial vectors we get much smoother spectral profiles than if we start the numerical optimization with uniformly distributed random numbers in $[0, 1]$, see Fig. 4 (right) with its highly oscillatory profiles. These results show that the reconstruction problem has many solutions, most of which are not chemically interpretable. For large data dimensions k and n there are too many degrees of freedom for the generator matrices. One way to reduce some of these degrees of freedom is to remove all data points from the inner polytopes that are not vertices. This means that the data is reduced to the so-called essential data [10, 14].

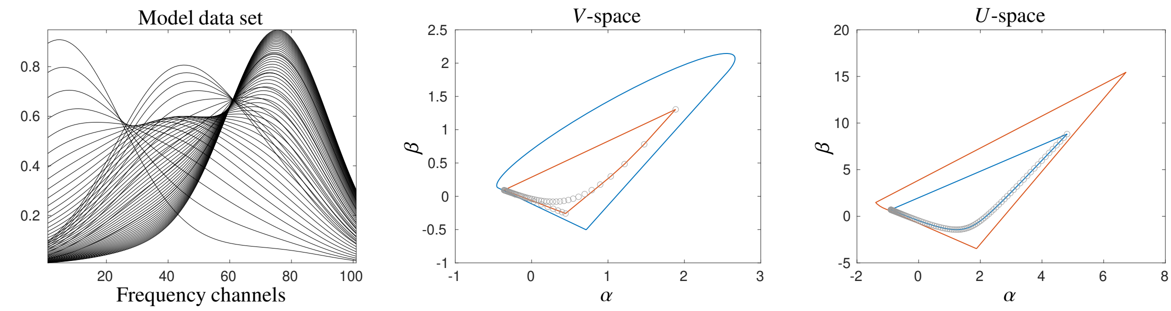


Figure 3: Left: Data of a three-component model system with rank$(D) = 3$. Center and right: The data points are marked by gray circles. The pairs of the dual inner and outer polygons are plotted in blue (or red) in the U- and the V-space.

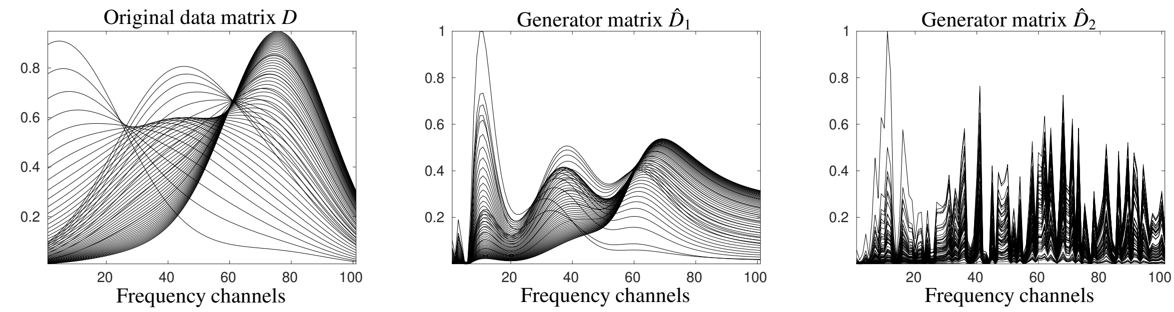


Figure 4: The original data matrix D (left) together with two generator matrices $\hat{D}_1$ and $\hat{D}_2$ (center and right). $\hat{D}_1$ was found by starting the iterative optimization with normalized all-ones vectors $(1, \ldots, 1)^T$ for u_1 and v_1. $\hat{D}_2$ was found by using normalized vectors of uniformly distributed random numbers between 0 and 1 as initialization. We used the MatLab optimizer `fminunc` for the computations.

3. Solution of Problem 1.2 by polytope adaptation from an initial reference problem

In simplified words, Problem 1.2 asks for the constructability of a generator matrix for freely designed polygons satisfying the inclusion $0 \in \mathcal{I} \subset \mathcal{F}$. It has to be stated, however, that a solution not always exists. This is illustrated by a simple example shown in Fig. 5.

3.1. A polytope adaptation algorithm

The key idea of how to solve Problem 1.2 is motivated by the procedure used when a spectral data matrix is augmented by additional spectra or when all spectra are considered on an extended spectral interval. Such data expansion amounts to adding more rows (spectra) or columns (spectral channels) to the spectral data matrix. These matrix expansion operations correspond to adding points in the U- and V-space and vice versa [2]. For given polytopes in the $(s-1)$-dimensional space, our polytope adaptation algorithm starts with a nonnegative initialization matrix with the rank s, which is then replaced by a matrix whose rows are filled with the spectral profiles represented by the vertices of the given (hand-drawn) inner polytope. A similar operation is then performed in the column space for the given dual inner polytope. The algorithm cannot be guaranteed to generate a matrix D that precisely reproduces the given polytopes, but numerical experiments for $s = 3$ indicate that the shape of the polygons is reproduced qualitatively.

We denote the two given target polytopes in the V-space by $\tilde{\mathcal{I}}_S$ and $\tilde{\mathcal{F}}_S$. These two polytopes determine the dual target polytopes $\tilde{\mathcal{I}}_C$ and $\tilde{\mathcal{F}}_C$ in the U-space. The steps of the polytope adaptation algorithm are as follows:

1. An initial nonnegative matrix D_0 with rank$(D_0) = s$ is chosen and the corresponding V-space is calculated by the SVD $D_0 = U_0\Sigma_0 V_0^T \in \mathbb{R}^{k_0\times n_0}$. Its polytopes can be very different from the target polytopes. (There is only one condition on the outer polytope of the initialization matrix, namely that it contains the given inner polytope $\tilde{\mathcal{I}}_S$ as otherwise the data points would represent profiles with negative entries. If this condition does not hold, then the given polytopes can be multiplied by a fixed positive constant less than 1, which will bring all points closer to the origin so that the inclusion condition can finally be satisfied.)
2. Let $y_j \in \mathbb{R}^{s-1}$, $j = 1, \ldots, k$, be the data points spanning the target polytope $\tilde{\mathcal{I}}_S$.
3. The k row vectors $(1, y_j^T)V_0^T$, $j = 1, \ldots, k$, are the spectral profiles represented by the data points of $\tilde{\mathcal{I}}_S$. These profiles are stored row by row in the intermediate matrix $D_1 \in \mathbb{R}^{k\times n_0}$.
4. An SVD $D_1 = U_1\Sigma_1 V_1^T \in \mathbb{R}^{k\times n_0}$ of D_1 is computed and then the V-space of $D_1^T = V_1\Sigma_1 U_1^T$ is considered. (Alternatively it is possible to consider the U-space of D_1, which is the same space up to scaling by Σ_1.)
5. Let $z_i \in \mathbb{R}^{s-1}$, $i = 1, \ldots, n$ be the data points of the dual target polytope $\tilde{\mathcal{I}}_C$.
6. The row vectors $(1, z_i^T)U_1^T$, $i = 1, \ldots, n$, are stored row by row in the matrix D^T resulting in the final matrix D.

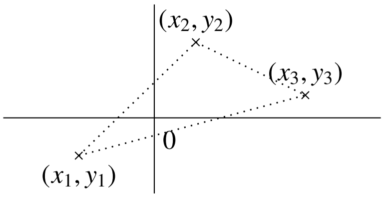


Figure 5: Three data points in the U-space spanning an inner polygon whose convex hull as marked by the dotted lines contains the origin. A feasible generator matrix must satisfy Eq. (5). The orthogonality of the second and third column of V requires $\vartheta := \mathrm{v}_{11}^2 x_1 y_1 + \mathrm{v}_{21}^2 x_2 y_2 + \mathrm{v}_{31}^2 x_3 y_3 = 0$. However, this condition cannot be satisfied since $\mathrm{v}_{i,1} > 0$ for $i = 1, 2, 3$ for the componentwise positive Perron vector v_1 and since for the shown choice of the coordinates it holds that $x_i y_i > 0$. So ϑ is strictly positive. No matrix V can be found and thus no generator matrix exists.

These steps are illustrated for an example data matrix in Fig. 6. The initial data matrix D_0 with rank s can be taken as

$$D_0 = \text{tridiag}(1, 3, 1) \in \mathbb{R}^{s\times s}.$$

This matrix is nonnegative. Its rank equals s because the Gerschgorin criterion limits its smallest eigenvalue from below by 1. Furthermore, $D_0 = D_0 I_{s\times s}$ is a nonnegative matrix factorization of D_0 where $I_{s\times s}$ is the $s \times s$ identity matrix.

This construction always yields a nonnegative matrix, because the data points on which the matrices are based are always inside of the outer polytopes and thus, the resulting spectra and concentration profiles that yield the data matrices are nonnegative. The inner and outer polytopes $\tilde{\mathcal{I}}_S$ and $\tilde{\mathcal{F}}_S$ can be chosen (almost) arbitrarily.

3.2. *Why does the polytope adaptation algorithm work?*

Next we show that the polytope adaptation algorithm maps the vertices of the initial polytopes (the data points underlying $\tilde{\mathcal{I}}_S$ and $\tilde{\mathcal{I}}_C$) to the vertices of the final polytopes of the generator matrix D. It is this vertex transformation property that allows us to consider the polytope adaptation algorithm to qualitatively preserve the original polytopes.

For this purpose, we consider the cones spanned by all linear combinations of the rows of a matrix with nonnegative coefficients. See the Definition Appendix A.2. The inner polytope $\mathcal{I}_S$ is the intersection of rowcone($U\Sigma$) with the $(s-1)$-dimensional affine hyperplane $\mathcal{H} = \{x \in \mathbb{R}^s : x_1 = 1\}$. This can also be described in a way that the linear combinations with positive coefficients of the data points underlying $\mathcal{I}_S$ (and augmented by a first component equal to 1) form the cone rowcone($U\Sigma$). In order to study these cones we analyze the related matrices.

The mathematical analysis of the steps of the polytope adaptation algorithm is as follows: Starting from an SVD of the initial matrix D_0, steps 3 and 4 of the polytope adaptation algorithm result in

$$\begin{aligned} D_0 &= U_0 \Sigma_0 V_0^T \\ D_1 &= \begin{pmatrix} 1, & y_1^T \\ & \vdots \\ 1, & y_k^T \end{pmatrix} V_0^T = (U_1 \Sigma_1 V_1^T) V_0^T = U_1 \Sigma_1 (V_0 V_1)^T \text{ where the data points } y_i \text{ define the target polytope } \tilde{\mathcal{I}}_S \end{aligned}$$

The key point is that $U_1\Sigma_1(V_0V_1)^T$ is an SVD of D_1, because orthonormal $V_0 \in \mathbb{R}^{n_0\times s}$ and orthogonal $V_1 \in \mathbb{R}^{s\times s}$ result in the orthonormal matrix V_0V_1. The further steps of the polygon adaptation algorithm result in

$$\begin{aligned} D_1^T &= (V_0 V_1) \Sigma_1 U_1^T \\ D^T &= \begin{pmatrix} 1, & z_1^T \\ & \vdots \\ 1, & z_n^T \end{pmatrix} U_1^T = (V_2 \Sigma_2 U_2^T) U_1^T = V_2 \Sigma_2 (U_1 U_2)^T \text{ where the } z_i \text{ define the target polytope } \tilde{\mathcal{I}}_C \end{aligned}$$

Again, $V_2\Sigma_2(U_1U_2)^T$ is an SVD of D^T, because orthonormal $U_1 \in \mathbb{R}^{k\times s}$ and orthogonal $U_2 \in \mathbb{R}^{s\times s}$ result in an orthonormal matrix U_1U_2.
These steps involve certain transformations of the data points of the target polytopes by the matrices from the different SVDs

$$\begin{pmatrix} 1, & y_1^T \\ & \vdots \\ 1, & y_k^T \end{pmatrix} = U_1 \Sigma_1 V_1^T, \text{ with } U_1 \in \mathbb{R}^{k\times s}, \Sigma_1,\ V_1 \in \mathbb{R}^{s\times s} \quad \text{and} \quad \begin{pmatrix} 1, & z_1^T \\ & \vdots \\ 1, & z_n^T \end{pmatrix} = V_2 \Sigma_2 U_2^T, \text{ with } V_2 \in \mathbb{R}^{n\times s}, \Sigma_2,\ U_2 \in \mathbb{R}^{s\times s}.$$

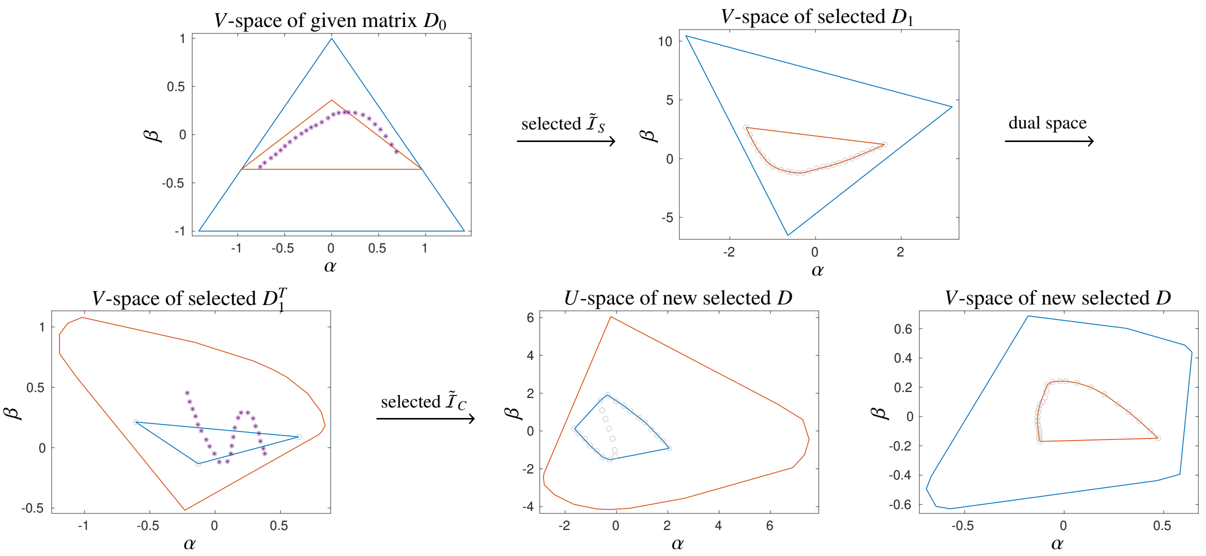


Figure 6: Illustration of the polytope adaptation algorithm. The chosen data points in the U-space and the V-space are marked with purple asterisks. First row: Left: V-space of the initialization matrix D_0. The outer polygon of D_0 is drawn in blue, the inner polygon in red, and the data points underlying $\tilde{\mathcal{I}}_S$ are the purple asterisks. Right: V-space of D_1, whose inner polygon (in red) approximately reproduces $\tilde{\mathcal{I}}_S$. The β-axis has a flipped orientation. This orientation has no effect on the profile representation, but is a consequence of a convention on how to orientate the singular vectors. Second row: Left: V-space of D_1^T with the data points underlying the dual polytope $\tilde{\mathcal{I}}_C$. Right: The U- and the V-spaces for the final generator matrix D together with the inner and outer polygons.

The SVD of the generator matrix D reads

$$D = U_1 U_2 \Sigma_2 V_2^T = U_3 \Sigma_3 V_3^T, \text{ where } U_3 = U_1 U_2,\ \Sigma_3 = \Sigma_2,\ V_3 = V_2.$$

Thus, the polytopes of D are based on the row-cones rowcone($U_3\Sigma_3$) (belonging to $\mathcal{I}_S$) and rowcone(V_3) (corresponding to $\mathcal{I}_C$). See Lemma Appendix A.3 for the relation between the edges of the row-cones and vertices of the polytopes. These cones result from the data points with

$$U_3\Sigma_3 = \begin{pmatrix} 1, & y_1^T \\ & \vdots \\ 1, & y_k^T \end{pmatrix} V_1 \Sigma_1^{-1} U_2 \Sigma_2, \qquad V_3 = \begin{pmatrix} 1, & z_1^T \\ & \vdots \\ 1, & z_n^T \end{pmatrix} U_2 \Sigma_2^{-1}.$$

We conclude that the data points undergo linear transformation with the two regular $s \times s$ matrices $\Sigma_1^{-1} U_2 \Sigma_2$ and Σ_2^{-1}. As shown in Lemma Appendix A.4, these transformations preserve the property of points of a polytope to be vertices. This is a qualitative property of the original polytopes that is preserved. Moreover, the vertices are the so-called essential data points, which finally determine the factor ambiguity.

3.3. Optimization-based construction of a generator matrix

An alternative to the geometry-inspired polytope adaptation algorithm, see Sec. 3.1, is an application of the optimization-based approach as explained in Sec. 2.2. Starting with any two polytopes $\mathcal{I}_S$ and $\mathcal{F}_S$ with $0 \in \mathcal{I}_S \subset \mathcal{F}_S$, duality relations determine the data points b_j [5, 9, 12]. Since it is not certain that the polytopes originate from a matrix, it is not guaranteed that a generator matrix exists, cf. with Fig. 5.

Numerically, we sometimes observed that the optimization stopped with large residuals and even negative entries of the computed generator matrix. This was observed mainly with global optimization algorithms and for randomly chosen data points without a stronger structure as can be expected for chemical data. It is also possible, that the polygons (or polytopes) of the resulting matrix are inverted along the coordinate axes, but this is only a consequence of the convention how to orientate the singular vectors in an SVD, see [13].

A useful application of the generator matrix construction is data sets with Borgen plots having unusual properties, for example MCR problems with a rank deficiency. Such examples are rather rare in experimental data sets, but provide interesting properties for the theoretical study of Borgen plots. We have chosen to illustrate this with two nested hexagons as the inner and outer polygons, see Fig. 7. By definition, the generator matrix of a two-dimensional Borgen plot is of rank 3. However, in the Borgen plot of Fig. 7 there is no nested triangle between the inner polygon and the outer polygon. This proves geometrically that there is no nonnegative factorization with rank-3 factors. The corresponding MCR problem suffers from a rank deficiency. This means that the generator matrix is of rank 3, but

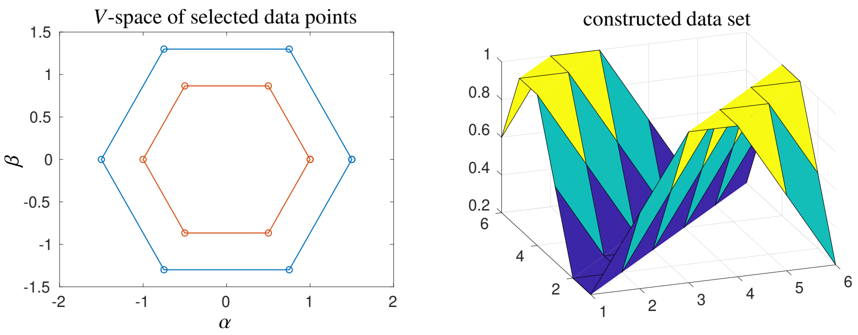


Figure 7: Left: Constructed inner and outer polygon in the V-space. The data points in the U-space are not needed due to duality. Right: The constructed data matrix corresponding to the V-space using the optimization approach.

cannot be factorized into two nonnegative matrices of rank 3. A similar Borgen plot was given in Fig. 2.6 in [4]. The reconstructed generator matrix D (using the optimizer `fminunc` and initializing with normalized ones vectors) has a similar structure to the data matrix in [4]. This supports the use of this approach to construct or adapt model problems with special properties.

3.4. Software availability

The software developed for the numerical experiments in this work is available from

`https://www.math.uni-rostock.de/facpack/Downloads.html`

The software is written in MatLab and can be operated via two simple GUIs. One for solving the problem via polygon adaptation and one using the optimization-based approach. The polygon adaptation GUI works just as described in Sec. 3.1. Starting from $D_0 = \text{tridiag}(1,3,1) \in \mathbb{R}^{3\times 3}$ the data points for $\tilde{\mathcal{I}}_S$ and $\tilde{\mathcal{I}}_C$ can be selected by hand and after confirming these, the final matrix D is computed. For the optimization-based approach, the user can load a rank-3 data matrix for which the inner and outer polygons are plotted. These polygons can then be modified by adding and removing any vertices, without breaking the $0 \in \mathcal{I} \subset \mathcal{F}$ inclusion relations. The associated spectral data matrix can then be recalculated with user-selectable initial estimates and for various optimization routines. Additionally, the user can start with a free geometric construction of the inner and outer polygons, for which the program will try to find a corresponding generator matrix.

4. Conclusion

Yes, it is possible to find a generator matrix underlying a Borgen plot and its generating polytopes. However, the problem is ill-posed in the sense that there are often many solutions and that the found numerical solution may show a strong dependence on the initial approximation. Another limitation is that the numerical algorithm does not necessarily terminate in a precise reconstruction of the given Borgen plot, but stops at a more or less precise approximation. Furthermore, the rows of the generator matrix are not guaranteed to be interpretable as the spectra of the underlying chemical reaction, since different generator matrices may result in the same Borgen plot. We hope that this work will stimulate scientific discussions among "Borgen-plot-experts" about questions what potential Borgen plot structures are possible and what happens when data points in existing data sets are changed, and thus to investigate properties of Borgen plots and the associated data matrices. The supplied software invites you to experiment.

Appendix A. Some properties of polytopes and cones

Lemma Appendix A.1. *Let $Y \in \mathbb{R}^{d\times d}$ be a regular matrix. The pairwise different vectors $z_i \in \mathbb{R}^d$, $i = 1, \ldots, m$, are the vertices of a polytope $\mathcal{P}$ if and only if Yz_i, $i = 1, \ldots, m$, are the vertices of a polytope $Y\mathcal{P}$.*

Proof. Let $w \in \mathcal{P}$ be a vertex of $\mathcal{P}$. Then w cannot be represented by convex combination of vertices $z_i \neq w$. Thus any representation

$$\sum_{i=1}^{m} c_i z_i - w = 0 \quad \text{with } c_i \geq 0 \text{ and } \sum_{i=1}^{m} c_i = 1$$

implies that all but one c_i are equal to zero. Since

$$Y\left(\sum_{i=1}^{m} c_i z_i - w\right) = \sum_{i=1}^{m} c_i Y z_i - Yw = 0$$

if and only if $\sum_{i=1}^m c_i z_i - w = 0$ completes the proof because, again, any convex combination representing Yw implies that all but one c_i are equal to zero. □

Next we define the row-cone which is spanned by all nonnegative linear combinations of the rows of a matrix.

Definition Appendix A.2 (see [2]). *Let a nonzero matrix $A \in \mathbb{R}^{k\times n}$ be given. The row-cone generated by the rows of A, namely* $\mathrm{rowcone}(A) \subseteq \mathbb{R}^{1\times n}$, *is defined as*

$$\mathrm{rowcone}(A) = \left\{ \sum_{i=1}^{k} \alpha_i A(i,:) : \textit{ for all } \alpha_i \geq 0 \right\} = \left\{ \sum_{i\in\mathcal{E}} \alpha_i A(i,:) : \textit{ for all } \alpha_i \geq 0 \right\},$$

where $\mathcal{E}$ denotes a smallest subset of the index set $\{1,\dots,k\}$. If all rows of A are pairwise different, then the subset $\mathcal{E}$ is unique. A vector $A(i,:)$ with $i \in \mathcal{E}$ lies on an edge of $\mathrm{rowcone}(A)$ *and the set of all positive multiples of $A(i,:)$ forms the edge.*

The next lemma shows that the vertices of the convex hull of the data points a_i according to (3) are related to the edges of $\mathrm{rowcone}(U\Sigma)$.

Lemma Appendix A.3. *A row vector $y \in \mathbb{R}_+^{1\times s}$ lies on an edge of* $\mathrm{rowcone}(U\Sigma)$ *if and only if the corresponding data point $(y(2:s))^T/y(1)$ is a vertex of $\mathcal{I}_S$.*

Proof. A vector $\tilde{y}$ lies on an edge of $\mathrm{rowcone}(U\Sigma)$ if and only if the representation

$$\tilde{y} = \sum_{i=1}^{k} \tilde{c}_i U\Sigma(i,:), \quad \tilde{c}_i \geq 0$$

implies that all but one of the $\tilde{c}_i$ are equal to zero. Within the affine plane of vectors whose first components are all equal to 1, this means that the representation

$$y = \sum_{i=1}^{k} c_i \frac{U\Sigma(i,:)}{U\Sigma(i,1)}, \quad \sum_{i=1}^{k} c_i = 1,\ c_i \geq 0,\ \text{with } y(1) = 1$$

implies that all but one of the c_i are equal to zero. Considering only the components $2,\dots s$ of the expansion of y reads

$$y(2:s) = \sum_{i=1}^{k} c_i \frac{U\Sigma(i,2:s)}{U\Sigma(i,1)} = \sum_{i=1}^{k} c_i a_i^T, \quad \sum_{i=1}^{k} c_i = 1,\ c_i \geq 0$$

where the vectors a_i are given by Eq. (3). This completes the proof. □

Lemma Appendix A.4. *A nonnegative matrix D and its positive multiple αD with $\alpha > 0$ have the same data vectors as given by Eq.* (3).

Proof. A singular value decomposition (SVD) $D = U\Sigma V^T$ of D leads to the SVD $(\alpha D) = U(\alpha\Sigma)V^T$ of αD with the same matrices of left and right singular vectors. The vectors $b_j = (V(j,2:s))^T/V(j,1)$ depend only on V, and in the data vectors $a_i = (((\alpha D)V)(i,2:s))^T/((\alpha D)V)(i,1)$ the scalar α can be reduced from the fraction. □

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