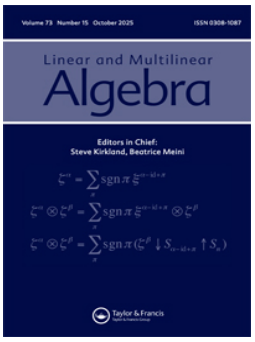

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On Sinkhorn's *DAD* theorem and the self-consistency equation in COSMO-based activity coefficient models

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ABSTRACT

In a 1966 paper, Sinkhorn proved that for any real square matrix A which has only positive entries there exists a uniquely determined real diagonal matrix D with positive diagonal entries such that $B := DAD$ is stochastic, i.e. all row sums of B are equal to 1. Moreover, Sinkhorn stated an iterative method for computing D. Nowadays, Sinkhorn's result and its variants are often referred to as *DAD* theorems. The purpose of this article is twofold. On the one hand, we give the link between Sinkhorn's *DAD* theorem and the self-consistency equation in COSMO-based activity coefficient models in chemical engineering. On the other hand, we give a new constructive proof of Sinkhorn's *DAD* theorem by using classical fixed-point theory. Hereby, the larger class of nonnegative matrices with positive diagonal is considered. Our proof uniformly provides convergence for a number of iterative methods for computing D. Some of them are used in practice although, to the best of our knowledge, a formal proof of convergence is missing.

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1. Introduction

In 1964 Sinkhorn [1] constructively proved his famous D_1AD_2 theorem which says that for each entrywise positive square matrix A there exist up to scalar multiples unique diagonal matrices D_1 and D_2 with positive diagonal entries such that D_1AD_2 is doubly stochastic, i.e. all row and column sums of D_1AD_2 are equal to 1. The famous Sinkhorn–Knopp algorithm [2] is a simple iterative method for computing D_1 and D_2. From Sinkhorn's result a vast literature on diagonal scalings of matrices with a wide variety of practical applications originated, see e.g. the survey article [3].

In 1966 Sinkhorn [4] also proved a similar but apparently lesser known *DAD* theorem which states that there exists a unique diagonal matrix D with positive diagonal entries such that DAD is stochastic, i.e. all row sums are equal to 1. Again, his proof is constructive, providing an iteration which converges to D. Soon after Sinkhorn's publication appeared, Brualdi, Parter, and Schneider [5, Thm. 7.6, Thm. 8.2, Cor. 7.7] presented a generalization

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of his result to a wide class of nonnegative matrices which particularly includes nonnegative matrices with positive diagonal. Their approach is not constructive and relies on Brouwer's fixed-point theorem. In contrast, the more recent paper by Labbé [6] from 2018 provides a constructive alternative approach for primitive nonnegative matrices with positive diagonal. In 1972 Csima [7] and 1974 Brualdi [8] focused on nonnegative symmetric matrices. Then, in 2009 Johnson and Reams [9, Thm. 1] proposed a different DAD algorithm and proved its convergence for positive A. We recommend this very readable paper and the references therein.

In general, it seems that D_1AD_2 algorithms are much better investigated in the literature than DAD algorithms, although their similarity is evident, especially because DAD algorithms produce a doubly stochastic scaling for symmetric matrices A. In this regard, we refer to Knight [10] and Knight and Ruiz [11] and the references therein. In [10, Thm. 4.4] an explicit expression for the linear convergence rate of the symmetric Sinkhorn-Knopp algorithm is stated for symmetric nonnegative fully indecomposable matrices in terms of the subdominant eigenvalue of DAD. In [11] a fast D_1AD_2 algorithm with quadratic convergence rate is presented which uses Newton's method and a preconditioned conjugate gradient method for efficiently solving the linear system in the Newton step. We also point to Aristodemo and Gemignani [12] where D_1AD_2 balancing is recasted to a nonlinear eigenproblem with eigenvector nonlinearity, and a self-consistent field iteration, a common approach in quantum chemistry (see e.g. [13]), is used to solve it. This significantly speeds up the Sinkhorn–Knopp iteration if D_1AD_2 has clustered dominant eigenvalues.

Recently [14–16], Sinkhorn's DAD scaling also emerged in physical chemistry and chemical engineering applications, appearing in the so-called *self-consistency equation* in conductor-like screening models (COSMO), introduced by Klamt [17]. Notably, similar equations have been in use since the 1980s in extensions of the quasi-chemical treatment [15, 18]. However, its connection to Sinkhorn's work [4] appears to be largely unrecognized in the technical literature on those models. In this work, we aim to bridge this gap. The self-consistency equation reads, see [15, Equation (10)]:

$$\Gamma_i = \left(\sum_{j=1}^{n} \theta_j \Gamma_j \Psi_{i,j}\right)^{-1} \quad \text{for } 1 \leqslant i \leqslant n. \tag{1}$$

Hereby, $\Gamma_i > 0$ is the *activity coefficient* of the fluid segment of type i, θ_j is the *surface area fraction* of segment type j, and $\Psi_{i,j}$ is the *contact Boltzmann factor* given by

$$\Psi_{i,j} := \exp\left(-\frac{u_{i,j}}{RT}\right) \quad \text{for } 1 \leqslant i,j \leqslant n, \tag{2}$$

where $u_{i,j} = u_{j,i}$ is the *contact formation energy* per mol of contacts for segment type pairs (i,j), R is the molar gas constant, and T is the temperature. See [15, 16] for details on the underlying chemical model. The surface area fractions θ_i are probabilities which sum up to 1, i.e. $\sum_{i=1}^{n} \theta_i = 1$. For the moment, let us assume that all θ_i are positive. The case of so-called *infinite dilution*, where some θ_i are zero, can easily be reduced to the nonzero case and is separately handled in Section 2.

Let $x := (\Gamma_1, \ldots, \Gamma_n)^\top$ be the column vector of the sought activity coefficients and let $D := \operatorname{diag}(x)$ be the diagonal matrix with diagonal entries $x_1 = \Gamma_1, \ldots, x_n = \Gamma_n$. The column vector of length n having 1 in each entry is denoted by $\mathbf{1}$ so that $D\mathbf{1} = x$.

The matrix $A := (a_{i,j})$ with

$$a_{i,j} := \theta_j \Psi_{i,j} \tag{3}$$

is positive, i.e. $a_{i,j} > 0$ for all $1 \leqslant i, j \leqslant n$. With this notation the self-consistency equation (1) reads $x = (Ax)^{-1}$ or equivalently

$$Ax = x^{-1} \tag{4}$$

where inversion is meant entrywise, i.e. $z^{-1} := (z_1^{-1}, \dots, z_n^{-1})^\top$ for a column vector $z = (z_1, \dots, z_n)^\top$ with positive entries z_i. Multiplying (4) from the left by D yields

$$DAD\mathbf{1} = DAx = Dx^{-1} = \mathbf{1}. \tag{5}$$

Thus, all row sums of DAD are equal to 1, i.e. DAD is stochastic. Therefore, Sinkhorn's result supplies existence and uniqueness of the diagonal matrix D and hence of the activity coefficients $\Gamma_1, \dots, \Gamma_n$ solving (1).

Possani and Soares [14] use an averaged successive substitution method for computing the activity coefficients $x = \Gamma$. This goes back to the work of Klamt [17]. To the best of our knowledge, a formal proof of convergence of this method does not exist in the literature. We will give such a proof by applying a classical fixed-point theorem of Edelstein [19] on contractive mappings, see Theorem 4.2 and Corollary 4.1. In particular, this provides a new constructive proof of Sinkhorn's DAD theorem.

The article is organized as follows. The short, preliminary Section 2 solves the case of infinite dilution. Section 3 states known algorithms for approximating the diagonal matrix D of Sinkhorn's DAD theorem. Precisely, these are the iterative schemes of Sinkhorn [4], Johnson and Reams [9], and the averaged successive substitution method [14]. Moreover, we include a Newton-type method recently proposed by Yan [20]. For all methods, short MATLAB implementations are provided.

Section 4 is the main part of this article containing our main new achievement. We present a unified proof of convergence for various DAD algorithms. In particular, this uniformly proves convergence of the Johnson-Reams method, the averaged substitution method, and the damped successive substitution used in Yan's method. The proof is formulated for nonnegative matrices A with positive diagonal. Note that Johnson and Reams [9, Thm. 1] only proved convergence of their method for positive A. Also primitivity of A as required by Labbé [6, Thm. 1] is not needed.

Finally, Section 5 contains a few low-dimensional examples of real chemical mixtures to illustrate numerically the convergence of the algorithms discussed in the previous sections. This might be particularly helpful for practitioners working in the area of COSMO-based activity coefficient models.

2. Infinite dilution

An important limiting case in COSMO-based models is so-called infinite dilution, in which some of the surface area fractions θ_j are zero. In such a case, let

$$J := \{j \in \{1, \dots, n\} \,:\, \theta_j = 0\}$$

be the index set collecting all these indices. The complementary set $K := \{1, \dots, n\} \backslash J$ is nonempty as $\sum_{i=1}^n \theta_i = 1$. For $j \in J$, the jth column of the matrix A in (3) is zero, and for $k \in K$ the kth column of A has solely nonzero entries. Let $A_K := (a_{i,j})_{i,j \in K}$ and $A_{J,K} := (a_{i,j})_{i \in J, j \in K}$. Then, A_K is a positive square matrix of order $|K| < n$ and $A_{J,K}$ is a positive matrix of size $|J| \times |K|$. According to Sinkhorn's theorem, let $\widetilde{D}$ be the diagonal matrix with positive diagonal y such that $\widetilde{D} A_K \widetilde{D}$ is stochastic. Set $z := [(A_{J,K})y]^{-1}$ and define $x \in \mathbb{R}^n$ by $x_K := y$ and $x_J := z$, whereby $x_L := (x_\ell)_{\ell \in L}$ denotes the subvector of x corresponding to a subset $L \subseteq \{1, \ldots, n\}$. Then, it is easy to verify that the matrix $D := \mathrm{diag}(x)$ fulfills $DAD\mathbf{1} = \mathbf{1}$. The auxiliary functions `zero_cols_reduce` and `zero_cols_resolve` in Listing 1 perform this reduction. Listing 6 shows how to call them before and after an arbitrary *DAD* algorithm.

```
function [AK,J,K] = zero_cols_reduce(A)
J = A(1,:)==0; % indices of zero columns
K = ~J;        % complementary indices
AK = A(K,K);   % principal submatrix of A with row and column indices K
end
function x = zero_cols_resolve(y,A,J,K)
x = zeros(size(A,1),1);
x(K) = y;
x(J) = (A(J,K)*y).^-1;
end
```

Listing 1. Infinite dilution.

As an example, consider a 4-by-4 matrix with zero second and fourth column:

$$A := \begin{bmatrix} 1/32 & 0 & 7/16 & 0 \\ 1/24 & 0 & 1/4 & 0 \\ 3/16 & 0 & 5/8 & 0 \\ 1/4 & 0 & 1/2 & 0 \end{bmatrix}.$$

Then, $J = \{2, 4\}$ and $K = \{1, 3\}$. Hence, $A_K = \begin{bmatrix} 1/32 & 7/16 \\ 3/16 & 5/8 \end{bmatrix}$ and $A_{J,K} = \begin{bmatrix} 1/24 & 1/4 \\ 1/4 & 1/2 \end{bmatrix}$.

For $y := [2, 1]^\top$ and $\widetilde{D} := \mathrm{diag}(y)$, the matrix $\widetilde{D} A_K \widetilde{D} = \begin{bmatrix} 1/8 & 7/8 \\ 3/8 & 5/8 \end{bmatrix}$ is stochastic. Now, define $z := [(A_{J,K})y]^{-1} = [3, 1]^\top$ and $x := [2, 3, 1, 1]^\top$ so that $x_K = y$ and $x_J = z$. Then, $D := \mathrm{diag}(x)$ scales A to a stochastic matrix:

$$DAD = \begin{bmatrix} 1/8 & 0 & 7/8 & 0 \\ 1/4 & 0 & 3/4 & 0 \\ 3/8 & 0 & 5/8 & 0 \\ 1/2 & 0 & 1/2 & 0 \end{bmatrix}.$$

Summing up, the case of infinite dilution can easily be reduced to a lower-dimensional case without infinite dilution, and all activity coefficients of the original, infinitely diluted mixture can be deduced from that. However, all algorithms stated in the next section, except Yan's method, can deal with infinite dilution without any reduction.

3. *DAD* algorithms

We start with introducing some simple notation which was partly already used in the introduction. For a real vector $x \in \mathbb{R}^n$ of length n and a real n-by-n matrix $M \in \mathbb{R}^{n \times n}$, the *maximum norm* of x and M are denoted by

$$\|x\| := \max_{1 \leqslant i \leqslant n} |x_i| \quad \text{and} \quad \|M\| := \max_{1 \leqslant i \leqslant n} \sum_{j=1}^{n} |M_{i,j}|.$$

For a function $f : \mathbb{R}_{>0} \to \mathbb{R}_{>0}$ and a vector $x = (x_1, \dots, x_n)^\top \in \mathbb{R}^n_{>0}$, we write

$$f(x) := (f(x_1), \dots, f(x_n))^\top$$

for the entrywise evaluation of f at x. For example,

$$1/x = x^{-1} = (x_1^{-1}, \dots, x_n^{-1})^\top, \quad \sqrt{x} = (\sqrt{x_1}, \dots, \sqrt{x_n})^\top,$$
$$\ln(x) = (\ln(x_1), \dots, \ln(x_n))^\top.$$

For $x, y \in \mathbb{R}^n_{>0}$, multiplication and division are also defined entrywise, i.e.

$$xy := (x_1y_1, \dots, x_ny_n)^\top \quad \text{and} \quad x/y := (x_1/y_1, \dots, x_n/y_n)^\top.$$

Also comparisons like $x \leqslant y$, $x < y$, $x \leqslant \alpha$, or $x < \alpha$ with $\alpha \in \mathbb{R}$ are meant entrywise, i.e. $x_i \leqslant y_i$, $x_i < y_i$, $x_i \leqslant \alpha$, or $x_i < \alpha$ for all $i \in \{1, \dots, n\}$, respectively.

Henceforth, $A = (a_{i,j})$ is an entrywise positive n-by-n matrix, if not stated otherwise.

3.1. Sinkhorn's iterations

In the proof of Lemma 1 in [4] two sequences $(x^{[k]})_{k \geqslant 0}$ and $(y^{[k]})_{k \geqslant 0}$ of positive vectors are iteratively constructed. Sinkhorn proves that $(x^{[k]}, y^{[k]})$ has a subsequence that converges to a pair of positive vectors $(\tilde{x}, \tilde{y})$ and that

$$D := \operatorname{diag}(x), \quad x := \sqrt{\tilde{y}_1/\tilde{x}_1} \cdot \tilde{x} \tag{6}$$

is the uniquely determined diagonal matrix with positive diagonal elements such that DAD is stochastic. We repeat Sinkhorn's original iteration S1 which is tailor-made for his proof but difficult to parse at first sight:

$$a := \min_{1 \leqslant i,j \leqslant n} a_{i,j}, \quad x_i^{[0]} := a^{-1/2}, \quad y_i^{[0]} := \left(\sum_j a_{i,j}\right)^{-1} a^{1/2}, \quad i = 1, \dots, n$$
$$x_i^{[k+1]} := \left(M^{[k]}\right)^{-1} a^{-1/2} \left(\rho_i^{[k]}\right)^{-1} x_i^{[k]}, \quad y_j^{[k+1]} := M^{[k]} a^{1/2} \left(\delta_j^{[k]}\right)^{-1} y_j^{[k]},$$
$$\rho_i^{[k]} := \sum_j x_i^{[k]} a_{i,j} y_j^{[k]}, \quad \delta_j^{[k]} := \sum_i \left(\rho_i^{[k]}\right)^{-1} x_i^{[k]} a_{j,i} y_j^{[k]},$$
$$M^{[k]} := \max_i \left(\rho_i^{[k]}\right)^{-1} x_i^{[k]}.$$

A closer inspection yields the following condensed formulation of S1:

$$\alpha := \sqrt{\min_{1 \leqslant i,j \leqslant n} a_{i,j}}, \quad x^{[0]} := \alpha^{-1}\mathbf{1}, \quad y^{[0]} := \left(Ax^{[0]}\right)^{-1}$$
$$z^{[k+1]} := \left(Ay^{[k]}\right)^{-1}, \quad x^{[k+1]} := \frac{z^{[k+1]}}{\alpha \|z^{[k+1]}\|}, \quad y^{[k+1]} := \left(Ax^{[k+1]}\right)^{-1}. \tag{7}$$

On the last page of [4] Sinkhorn states the following simplified, different iteration S2 which skips the scaling of the iterates by α and $\|z^{[k+1]}\|$:

$$x^{[0]} := \mathbf{1}, \quad y^{[0]} := \left(Ax^{[0]}\right)^{-1}$$
$$x^{[k+1]} := \left(Ay^{[k]}\right)^{-1}, \quad y^{[k+1]} := \left(Ax^{[k+1]}\right)^{-1}. \tag{8}$$

After termination of the iteration, the final result is again given by (6).

Listing 2 shows a MATLAB implementation of Sinkhorn's iterations. The first input argument `A` is the matrix. The second input argument `maxiter` is the maximum number of iterations. If the maximum entrywise relative distance of two consecutive iterates is smaller than the tolerance specified by the third input argument `tol`, then the iteration ends before reaching `maxiter`. The last input argument `s` allows to switch between S1 (`s`$:=1$) and S2 (`s`$:=2$). The first return argument `x` contains the diagonal of the sought diagonal matrix D. The second return argument `k` is the number of iterations that have been carried out. Let us finally recall and point out clearly that Sinkhorn only proved convergence of a subsequence of $(x^{[k]}, y^{[k]})$, However, by numerical evidence, the sequence $(x^{[k]}, y^{[k]})$ itself always converges.

```
function [x,k] = sinkhorn(A,maxiter,tol,s)
n = size(A,1); % dimension, matrix order
x = ones(n,1); % initialize x-values with all-ones vector
if s==1 % Sinnkhorn's algorithm S1
    K = A(1,:) > 0;
    if all(K)
        alpha = sqrt(min(A(:))); % minimum nonzero entry of A
    else % infinite dilution
        AK = A(K,K); alpha = sqrt(min(AK(:)));
    end
    x = alpha*x; % initialize x-values
end
y = (A*x).^-1;    % initialize y-values
k = 0;            % initialize iteration counter
t = tol + 1;
while t > tol & k < maxiter
    x_ = (A*x).^-1; % new x-values
    if s==1 % Sinnkhorn's algorithm S1
        x_ = x_/(alpha*norm(x_,'inf')); % new x-values
    end
    y_ = (A*x_).^-1;                % new y-values
    tx = norm((x-x_)./x,'inf');     % relative x-deviation
    ty = norm((y-y_)./y,'inf');     % relative y-deviation
    t = max(tx,ty);                 % overall maximum norm
    x = x_;                         % update old x-values for next loop
    y = y_;                         % update old y-values for next loop
    k = k+1;                        % increment iteration counter
end
x = sqrt(y(1)/x(1))*x; % final result
```

Listing 2. Sinkhorn's iterations S1 and S2.

3.2. Johnson-Reams iteration

Johnson and Reams [9] state in the proof of their Theorem 1 a new *DAD* scaling method which we abbreviate by JR. They consider diagonal matrices $D^{[k]} = \mathrm{diag}(x^{[k]})$ with positive diagonal vectors $x^{[k]}$ as defined in (9) and prove that these $D^{[k]}$ converge to a diagonal matrix D such that DAD is stochastic. The diagonal iteration is

$$x^{[0]} := \mathbf{1}, \quad x^{[k+1]} := \left(x^{[k]} / \left(Ax^{[k]}\right)\right)^{1/2}. \tag{9}$$

For an unknown reason, Johnson and Reams do not explicitly state this simple formula for their method. Listing 3 implements the Johnson-Reams iteration. Input and output arguments have the same meaning as in Sinkhorn's iterations in Listing 2.

```
function [x,k] = JR(A,maxiter,tol)
n = size(A,1); x = ones(n,1);  % initialize solution with all—ones vector
k = 0; t = tol + 1;
while t > tol & k < maxiter
    x_ = A*x;
    x_ = sqrt(x./x_);          % geometric mean of old and new values
    t = norm((x—x_)./x,'inf'); % relative deviation
    x = x_; k = k+1;
end
```

Listing 3. Johnson-Reams iteration.

3.3. *Averaged substitution method*

Possani and Soares [14, Fig. 4, Alg. 1] state an averaged successive substitution method (AVS) for computing the activity coefficients $x = \Gamma$ that solve the self-consistency equation (1). This goes back to the original work of Klamt [17]:

$$x^{[0]} := \mathbf{1}, \quad x^{[k+1]} := \frac{1}{2}\left(x^{[k]} + \left(Ax^{[k]}\right)^{-1}\right). \tag{10}$$

Possani and Soares report that taking the average of the old vector $x^{[k]}$ and the new vector $(Ax^{[k]})^{-1}$ is done to prevent oscillations but can slow down convergence.

Comparing with Sinkhorn's iteration, the occurrence of such oscillations between the separate sequences $x^{[k]}$ and $y^{[k]}$ in (7) and (8), respectively, is not surprising. There, an averaging is only done once by (6) at the very end of the iteration. Comparison of (10) and (9) shows that the only difference between AVS and JR is that AVS uses the arithmetic mean for averaging while JR uses the geometric mean. An implementation of AVS is given in Listing 4.

```
function [x,k] = AVS(A,maxiter,tol)
n = size(A,1); x = ones(n,1);  % initialize solution with all—ones vector
k = 0; t = tol + 1;
while t > tol & k < maxiter
    x_ = (A*x).^—1;            % new values
    x_ = (x+x_)/2;             % arithmetic mean of old and new values
    t = norm((x—x_)./x,'inf'); % relative deviation
    x = x_; k = k+1;
end
```

Listing 4. Averaged successive substitution method.

For $n = 1$, we have $A = a > 0$ and (10) becomes $x^{[k+1]} = \frac{1}{2}(x^{[k]} + a^{-1}/x^{[k]})$ which is Heron's method (also called Babylonian method) for approximating the square root of a^{-1}. Thus, AVS can be seen as a generalization of Heron's method to higher dimensions for approximating Sinkhorn's diagonal matrix D making DAD stochastic. To the best of our knowledge, a proof of convergence of AVS for general n has not been given yet. We will give such a proof in a more general form, valid for a wider class of means including the arithmetic and the geometric mean, in Section 4.

3.4. Yan's method

Recently, Yan [20] proposed a specific Newton-type method, which we abbreviate by Y, for solving the self-consistency equation (1). The method is comparable to the fast balancing algorithm by Knight and Ruiz [11] for symmetric A which they implemented as a MATLAB function `bnewt` in the Appendix of [11]. Yan's method takes advantage of the special structure of the matrix $A = \Psi S$ defined in (3), where $\Psi = (\Psi_{i,j})$ is the matrix of the contact Boltzmann factors defined in (2) and

$$S := \operatorname{diag}(\theta) = \operatorname{diag}(\theta_1, \dots, \theta_n)$$

is the diagonal matrix with the surface area fractions on its diagonal. Note that Ψ is symmetric but A is in general not. However,

$$H := SA = S\Psi S$$

is symmetric again. In order to maintain symmetry, the transformed self-consistency equation (5) can be multiplied from the left by S, which gives

$$x(Hx) = DHx = DSAx = SDAx = S\mathbf{1} = \theta.$$

Recall that $D = \operatorname{diag}(x)$. Thus, solving the self-consistency equation is equivalent to finding an entrywise positive root of the function

$$f : \mathbb{R}^n \to \mathbb{R}^n, \quad x \mapsto x(Hx) - \theta.$$

The Jacobian $J(x)$ of f at $x \in \mathbb{R}^n$ readily computes as

$$J(x) = \operatorname{diag}(x)H + \operatorname{diag}(Hx).$$

The update term Δx of Newton's method applied to f calculates as

$$J(x)\,\Delta x = -f(x).$$

Multiplying both sides from the left by x^{-1} yields

$$\big(H + \operatorname{diag}((Hx)/x)\big)\ \Delta x = \theta/x - Hx.$$

Abbreviating $\widehat{H} := -(H + \operatorname{diag}((Hx)/x))$ and $g := \theta/x - Hx$, we get

$$\widehat{H}\Delta x + g = 0. \tag{11}$$

This is exactly Equation (55) of Yan's method [20] for solving the self-consistency equation. As Yan [20] points out, the benefit is that the so-called modified Hessian $\widehat{H}$ is symmetric and can even be shown to be negative definite so that convergence of the Newton method can be assured. Yan [20], Sec. 4., employs a Cholesky factorization to solve the linear system (11) for Δx. In contrast, Knight and Ruiz [11], use a more efficient conjugate gradient method for solving the linear system in the Newton step.[1]

Listing 5 implements Yan's method along the lines stated in steps (1) to (9) in Section 4 of [20]. These steps are indicated as comments in the code.

```
function [x,k] = Y(A,maxiter,tol)
n = size(A,1);    % dimension, matrix order
k = 0;            % initialize iteration counter
t = tol + 1;
% step (1) is dropped since temperature dependence is not considered
% step (2): retrieve theta from A
theta = 1./sum(A./A',2);
% step (3): initialize solution with all-ones vector
x = ones(n,1);
% step(4)[optional]: perform several damped successive iterations
%                    to get a better starting point x
damped_iter = 3; % number of successive damped iterations
omega = 0.2;     % damping factor; omega = 0.5 would be the AVS method
for i = 1:damped_iter
    x_ = (A*x).^-1; x_ = (1-omega)*x_ + omega*x; x = x_;
end
H = theta.*A;
Q = theta'*log(x)-0.5*x'*H*x+0.5; % objective function to be maximized
opts.SYM = true;     % options for MATLAB's fast built-in function linsolve
opts.POSDEF = true; % to invoke Cholesky factorization
H_ = H; h = diag(H);
ix = 1:n+1:n^2;      % index of the diagonal
while t > tol & k < maxiter
    % step (5): Newton iteration
    H_(ix) = h+(H*x)./x;          % H_ = H+diag((H*x)./x) neg.modified Hessian
    g = theta./x-H*x;             % right-hand side of Newton iteration
    dx = linsolve(H_,g,opts);     % solve linear system H_ * dx = g
    x_ = x+dx;                    % Newton update
    % step (6): scale down dx if x_ is not entrywise positive
    idx = x_ <= 0;
    if any(idx)
       y = x(idx); dy = dx(idx); c = 0.5*min(y./-dy); x_ = x + c*dx;
    end
    % step (7): reduce dx by half if Q does not increase
    Q_ = theta'*log(x_)-0.5*x_'*H*x_+0.5;
    while Q_ < Q
          dx = dx/2; x_ = x+dx; Q_ = theta'*log(x_)-0.5*x_'*H*x_+0.5;
    end
    % step (8): check for convergence and update old values for next loop
    t = norm((x-x_)./x,'inf'); % maximum norm of relative iterate deviation
    x = x_;                       % update old values for next loop
    k = k+1;                      % increment iteration counter
    % step (9) is dropped since temperature dependence is not considered
end
k = k + damped_iter; % total iteration count
```

Listing 5. Yan's method.

4. Convergence analysis

We start with some simple definitions.

Definition 4.1: A function $f : \mathbb{R}^n_{>0} \to \mathbb{R}^m_{>0}$ is called

(a) *homogeneous* if $f(\alpha x) = \alpha f(x)$ for all $\alpha > 0$ and all $x \in \mathbb{R}^n_{>0}$,
(b) *strictly monotone* if $f(x) < f(y)$ for all $x, y \in \mathbb{R}^n_{>0}$ with $x \leqslant y$ and $x \neq y$,
(c) *diagonally monotone* if $m = n$ and $f(x) \leqslant f(y)$ for all $x, y \in \mathbb{R}^n_{>0}$ with $x \leqslant y$ whereby $[f(x)]_i < [f(y)]_i$ for all $i \in \{1, \dots, n\}$ with $x_i < y_i$,
(d) *diagonally bounded* if $m = n$ and if there are $\mu, \nu > 0$ such that $\mu x \leqslant f(x) \leqslant \nu \|x\|$ for all $x \in \mathbb{R}^n_{>0}$.

Recall that the inequality in (d) means $\mu x_i \leqslant [f(x)]_i \leqslant \nu \|x\|$ for $i = 1, \dots, m$. Since $\|x\| > 0$ is attained for some i, this also means $\mu \leqslant \nu$.

Example 4.1: Let $A = (a_{i,j})$ be a nonnegative n-by-n matrix with positive diagonal. Then, $f(x) := Ax, x \in \mathbb{R}^n_{>0}$, is homogeneous, diagonally monotone, and diagonally bounded. The constants in Definition 4.1(d) can be taken as $\mu := \min_{1 \leqslant i \leqslant n} a_{i,i}$ and $\nu := \|A\|$.

Definition 4.2: A homogeneous and strictly monotone function $g : \mathbb{R}^2_{>0} \to \mathbb{R}_{>0}$ with $g(x, x) = x$ for all $x > 0$ is called a *mean*.

Example 4.2: For each $\omega \in (0, 1)$, the functions

$$g(x, y) := \omega x + (1 - \omega) y \quad \text{and} \quad h(x, y) := x^{\omega} y^{1-\omega}$$

are means. For $\omega = 1/2$, these are the arithmetic and geometric mean.

For a function $g : \mathbb{R}^2_{>0} \to \mathbb{R}_{>0}$, $x, y \in \mathbb{R}^n_{>0}$, and $\alpha \in \mathbb{R}_{>0}$, we abbreviate

$$g(x, y) := (g(x_1, y_1), \dots, g(x_n, y_n))^\top, \quad g(x, \alpha) := g(x, \alpha \cdot \mathbf{1}), \quad g(\alpha, y) := g(\alpha \cdot \mathbf{1}, y).$$

Lemma 4.1: *Let $f : \mathbb{R}^n_{>0} \to \mathbb{R}^n_{>0}$ be diagonally bounded, let $g(x, y) := \sqrt{xy}$ be the geometric mean, and define*

$$F : \mathbb{R}^n_{>0} \to \mathbb{R}^n_{>0}, \quad x \mapsto g(x, 1/f(x)).$$

Furthermore, let $x^{[0]} \in \mathbb{R}^n_{>0}$ and $x^{[k]} := F^k(x^{[0]})$, $k \in \mathbb{N}$, be the fixed-point iteration of F starting at $x^{[0]}$. There are $a, b \in \mathbb{R}_{>0}$ with $a \leqslant b$ such that $x^{[k]} \in [a, b]^n$ for all $k \in \mathbb{N}_0$. In particular, the sequence $(x^{[k]})$ is bounded and entrywise bounded away from zero.

Proof: Let μ, ν be as in Definition 4.1(d) and define

$$c := \max\{1, \mu^{-1}, \nu, \|(x^{[0]})^2\|, \|(x^{[0]})^{-2}\|\}.$$

Then,

$$a := c^{-1/2} \leqslant x^{[0]} \leqslant c^{1/2} =: b \tag{12}$$

$$c^{-1} \leqslant \mu \leqslant \nu \leqslant c. \tag{13}$$

We will now prove by induction that

$$\|x^{[k]}\| \in [a, b] \quad \text{for all } k \in \mathbb{N}_0. \tag{14}$$

For $k = 0$, this is clear by (12). Let $k \geqslant 1$ and assume $\|x^{[k-1]}\| \in [a, b]$. By strict monotonicity of g, Definition 4.1(d), and (13) it follows that

$$\|x^{[k]}\| \geqslant x^{[k]} = g\left(x^{[k-1]}, \frac{1}{f(x^{[k-1]})}\right) \geqslant g\left(x^{[k-1]}, \frac{1}{\nu\|x^{[k-1]}\|}\right) \geqslant g\left(x^{[k-1]}, \frac{1}{c\|x^{[k-1]}\|}\right).$$

Since $\|x^{[k-1]}\|$ is attained in one component of $x^{[k-1]}$, this implies

$$\|x^{[k]}\| \geqslant g\left(\|x^{[k-1]}\|, \frac{1}{c\|x^{[k-1]}\|}\right) = c^{-1/2} = a.$$

Since f is diagonally bounded, Definition 4.1(d) yields $f(x) \geqslant \mu x$ for all $x \in \mathbb{R}^n_{>0}$. Using also $\mu^{-1} \leqslant c$ according to (13), we get

$$x^{[k]} = g\left(x^{[k-1]}, \frac{1}{f(x^{[k-1]})}\right) \leqslant g\left(x^{[k-1]}, \frac{1}{\mu x^{[k-1]}}\right) \leqslant g\left(x^{[k-1]}, \frac{c}{x^{[k-1]}}\right) = \sqrt{c} = b.$$

Hence, $\|x^{[k]}\| \leqslant b$. This finishes the proof of (14). Now, define $\widehat{a} := \min\{a, 1/(cb)\}$. We will prove by induction that

$$\widehat{a} \leqslant x^{[k]} \quad \text{for all } k \in \mathbb{N}_0. \tag{15}$$

For $k = 0$, this is clear by (12) as $\widehat{a} \leqslant a$. Let $k \geqslant 1$ and assume $\widehat{a} \leqslant x^{[k-1]}$. Using also $\|x^{[k-1]}\| \leqslant b$ according to (14), we estimate

$$x^{[k]} = g\left(x^{[k-1]}, \frac{1}{f(x^{[k-1]})}\right) \geqslant g\left(x^{[k-1]}, \frac{1}{\nu\|x^{[k-1]}\|}\right) \geqslant g\left(\widehat{a}, \frac{1}{cb}\right) \geqslant g(\widehat{a}, \widehat{a}) = \widehat{a}.$$

Thus, (14) and (15) give $x^{[k]} \in [\widehat{a}, b]^n$ for all $k \in \mathbb{N}_0$ which proves the assertion. ∎

Next, the Thompson metric [21] on $\mathbb{R}^n_{>0}$ is defined by

$$d(x, y) := \ln\left(\max\{\|x/y\|, \|y/x\|\}\right) = \|\ln(x) - \ln(y)\| \quad \text{for } x, y \in \mathbb{R}^n_{>0}. \tag{16}$$

Since $\ln : \mathbb{R}_{>0} \to \mathbb{R}$ is a homeomorphism, the topology induced by Thompson's metric on $\mathbb{R}^n_{>0}$ is the usual Euclidean one so that we do not have to distinguish between them regarding topological properties like convergence, openness, compactness, etc.

Lemma 4.2: *Let f be a homogeneous and diagonally monotone function on $\mathbb{R}^n_{>0}$ and let $g : \mathbb{R}^2_{>0} \to \mathbb{R}_{>0}$ be a mean. Then, the function $F(x) := g(x, 1/f(x))$ is contractive w.r.t. the Thompson metric, i.e. $d(F(x), F(y)) < d(x, y)$ for all $x, y \in \mathbb{R}^n_{>0}$ with $x \neq y$.*

Proof: Pick $x, y \in \mathbb{R}^n_{>0}$ and abbreviate $\alpha := \|x/y\|$ and $\beta := \|y/x\|$. Then,

$$y/\beta \leqslant y/(y/x) = x = (x/y)y \leqslant \alpha y.$$

Homogeneity, and monotonicity of f and g yield

$$\begin{aligned} F(x) &= g(x, 1/f(x)) \leqslant g(\alpha y, 1/f(y/\beta)) && (17)\\ &= g(\alpha y, \beta/f(y)) \leqslant g(\max\{\alpha, \beta\}y, \max\{\alpha, \beta\}/f(y)) && (18)\\ &= \max\{\alpha, \beta\}g(y, 1/f(y)) = \max\{\alpha, \beta\}F(y). \end{aligned}$$

Note carefully that equality in (17) in one single component $i \in \{1, \ldots, n\}$ implies $x_i = \alpha y_i$ and $[f(x)]_i = [f(y/\beta)]_i$ by strict monotonicity of g. The latter necessitates $x_i = y_i/\beta$ due to diagonal monotonicity of f and the former gives $\alpha y_i = x_i = y_i/\beta$ so that $\alpha = \beta^{-1}$. Likewise, equality in (18) in one single component already implies $\alpha = \beta = \max\{\alpha, \beta\}$ due to strict monotonicity of g. Thus, we conclude

$$\|F(x)/F(y)\| \leqslant \max\{\alpha, \beta\}$$

and equality holds if and only if $\beta^{-1} = \alpha = \beta$ which means $\alpha = \beta = 1$ and hence $x = y$. By symmetry, $\|F(y)/F(x)\| \leqslant \max\{\alpha, \beta\}$ and equality also implies $x = y$. Together, this gives

$$\max\{\|F(x)/F(y)\|, \|F(y)/F(x)\|\} \leqslant \max\{\alpha, \beta\}.$$

Taking logarithms yields $d(F(x), F(y)) \leqslant d(x, y)$ and equality implies $x = y$. ■

The following theorem of Edelstein [19, Theorem 1 and Remark 3.2] is a classical result in fixed-point theory, generalizing Banach's fixed-point theorem.

Theorem 4.1 (Edelstein): *Let (X, d) be a metric space and f a contractive self-mapping of X, i.e. $f(X) \subseteq X$ and $d(f(x), f(y)) < d(x, y)$ for all distinct $x, y \in X$. Suppose that there exists an $x \in X$ such that the sequence of iterates $(f^k(x))_{k \in \mathbb{N}}$ has a subsequence $(f^{k_i}(x))_{i \in \mathbb{N}}$ that converges to a point $\xi \in X$. Then, ξ is a unique fixed point of f and $\xi = \lim_{k \to \infty} f^k(x)$.*

We will now apply Edelstein's theorem to our specific setting.

Theorem 4.2: *Let f be a homogeneous, diagonally bounded, and diagonally monotone function on $\mathbb{R}^n_{>0}$ and let $g : \mathbb{R}^2_{>0} \to \mathbb{R}_{>0}$ be a mean. Then, $F(x) := g(x, 1/f(x))$ has a unique fixed point $z \in \mathbb{R}^n_{>0}$ and for each $x \in \mathbb{R}^n_{>0}$ it holds that $\lim_{k \to \infty} F^k(x) = z$. The fixed point fulfills $f(z) = z^{-1}$ and is unique w.r.t. this property.*

Proof: By Lemma 4.2, F is contractive w.r.t. the Thompson metric. Pick $x \in \mathbb{R}^n_{>0}$. First, we consider the case where g is the geometric mean. By Lemma 4.1, the sequence of iterates $(F^k(x))_{k\in\mathbb{N}_0}$ is bounded and entrywise bounded away from zero. Thus, by the Bolzano–Weierstrass theorem, it has a convergent subsequence with limit $z \in \mathbb{R}^n_{>0}$. Theorem 4.1 applied to F yields that z is the unique fixed point of F and $z = \lim_{k\to\infty} F^k(x)$. Since $g(y,y) = y$ for all $y \in \mathbb{R}^n_{>0}$ and since g is strictly monotone, the equality $g(z,z) = z = F(z) = g(z, f(z)^{-1})$ is equivalent to $z = f(z)^{-1}$, i.e. $f(z) = z^{-1}$. This finishes the proof for the geometric mean.

Next, we consider an arbitrary mean g. By the previous case, we already know that there is a unique $z \in \mathbb{R}^n_{>0}$ such that $f(z) = z^{-1}$. Clearly, z remains a fixed point of the newly defined F as $F(z) = g(z, f(z)^{-1}) = g(z,z) = z$. Thus,

$$d(z, F^k(x)) = d(F^k(z), F^k(x)) \leqslant d(z,x) =: r \quad \text{for all } k \in \mathbb{N}_0.$$

By definition of the Thompson metric, see (16), this means

$$0 < a := 1/(\|z^{-1}\| e^r) \leqslant z/e^r \leqslant F^k(x) \leqslant z\, e^r \leqslant \|z\| e^r =: b \quad \text{for all } k \in \mathbb{N}_0.$$

Hence, the sequence of iterates $(F^k(x))_{k\in\mathbb{N}_0}$ is contained in $[a,b]^n$ and is therefore bounded and entrywise bounded away from zero. Now, the assertion follows word by word as in the previous case. ■

With hindsight, the existence of a convergent subsequence of the fixed point iteration $F^k(x)$ immediately followed from Lemma 4.1 in case of the geometric mean. With this requirement of Edelstein's fixed point theorem at hand, a $z \in \mathbb{R}^n_{>0}$ fulfilling $z = f(z)^{-1}$ was found. Once, existence of such a z was established, it was a posteriori easy to verify convergence of a subsequence of the fixed point iteration $F^k(x)$ for general means. It seems not obvious how to prove this fact directly without the detour via the geometric mean because the proof of Lemma 4.1 strongly relies on specific, favorable properties of the geometric mean which are not available for general means.

Corollary 4.1: *For every entrywise nonnegative n-by-n matrix A with positive diagonal entries, there is a unique $x \in \mathbb{R}^n_{>0}$ such that $Ax = x^{-1}$, i.e. DAD is stochastic where* $D := \operatorname{diag}(x)$. *The Johnson-Reams method* (9), *the averaged substitution method* (10), *as well as the damped iteration in step* (4) *of Yan's method in Listing* 5 *converge to x started with any* $x^{[0]} \in \mathbb{R}^n_{>0}$.

Proof: This follows from Theorem 4.2 with f as in Example 4.1 and g the geometric mean for JR, g the arithmetic mean for AVS, and $g(x,y) := 0.2x + 0.8y$ for Yan's damped iteration. ■

By numerical evidence, it seems that the Johnson-Reams and the averaged substitution method even work for wider classes of entrywise nonnegative matrices A for which there exists a unique diagonal matrix D with positive diagonal entries such that DAD is stochastic. A large but not complete such class is stated in [5, Thm. 8.2]. Namely, these are all nonnegative n-by-n matrices A with nonzero rows which either have a positive diagonal or which are permutationally similar to a matrix of the form

$$\begin{array}{c} \\ k \rightarrow \\ \end{array} \begin{array}{c} \qquad\qquad\qquad\quad k \\ \qquad\qquad\qquad\quad \downarrow \\ \left(\begin{array}{c|c|c|c} \mathbf{0_{r\times r}} & * & \mathbf{v} & * \\ \hline * & \cdots & \cdots & * \\ \hline \mathbf{0_{1\times r}} & * & \cdots & * \\ \hline * & \cdots & \cdots & * \end{array}\right) \end{array} \tag{19}$$

where $1 \leqslant r \leqslant n-1$, $r+1 \leqslant k \leqslant n$, and $\mathbf{v} \in \mathbb{R}^r_{\geqslant 0}$ is nonzero. Hereby $\mathbf{0_{r\times r}}$ and $\mathbf{0_{1\times r}}$ denote the r-by-r zero matrix and the zero row vector of length r, respectively.

It remains an open question whether the presented convergence analysis can be extended to matrices with nonzero rows and zero pattern (19).

5. Examples

In what follows, the algorithms discussed in the previous sections are applied to three low-dimensional examples which correspond to real chemical mixtures. This is just to illustrate numerically the convergence of those algorithms and might be helpful for practitioners working on COSMO-based activity coefficient models. The executable MATLAB frame program in Listing 6 can be used to produce the stated results by replacing the random matrix A in line 4 by the ones from the examples.

```
1  clc, n = 5; e = ones(n,1); maxiter = 500; tol = 1e-12;
2  Psi = rand(n); Psi = Psi + Psi'; theta = rand(n,1);
3  theta(randi([1,n],[min(2,n-1),1])) = 0; theta = theta/sum(theta);
4  A = Psi.*theta', iter = zeros(5,1); res = zeros(5,1); x_ = zeros(n,5);
5  for z = 1:5
6    if any(theta==0) % check infinite dilution
7      A_ = A; [A,J,K] = zero_cols_reduce(A);
8    end
9    switch z
10     case 1 % Sinkhorn algorithm S1
11       [x,k] = sinkhorn(A,maxiter,tol,1);
12     case 2 % Sinkhorn algorithm S2
13       [x,k] = sinkhorn(A,maxiter,tol,2);
14     case 3 % Johnson-Reams algorithm
15       [x,k] = JR(A,maxiter,tol);
16     case 4 % averaged successive substitution method
17       [x,k] = AVS(A,maxiter,tol);
18     case 5 % Yan's method
19       [x,k] = Y(A,maxiter,tol);
20   end
21   if any(theta==0)
22     A = A_; x = zero_cols_resolve(x,A,J,K);
23   end
24   r = x.*(A*x)-e; % residual r = (DAD)e-e, D = diag(x)
25   res(z) = norm(r,'inf'); iter(z) = k; x_(:,z) = x;
26 end
27 format short; R = erase(strcat("x_",num2str((1:n).'))," ");
28 X = array2table(x_,'VariableNames',{'S1','S2','JR','AVS','Y'},...
29     'RowNames',R); disp(X)
30 T = array2table([res,iter],'VariableNames',{'residual','iterations'},...
31     'RowNames',{'S1','S2','JR','AVS','Y'}); disp(T)
```

Listing 6. Frame program for *DAD* algorithms.

Example 1 is an equimolar mixture of five compounds with equal surface area fractions $\theta_j = 0.2$, $j = 1, \ldots, 5$. Originally proposed by [22], this is an extremely nonideal system with both positive (repulsive) and negative (attractive) interaction energies. The corresponding matrix $A = 0.2\Psi$ is symmetric as Ψ is, see (2), (3):

$$A = \begin{bmatrix} 0.2 & 0.44814116 & 0.08925759 & 0.16346839 & 0.05962834 \\ 0.44814116 & 0.2 & 0.13360957 & 0.67082194 & 0.18449794 \\ 0.08925759 & 0.13360957 & 0.2 & 0.08925759 & 0.05962834 \\ 0.16346839 & 0.67082194 & 0.08925759 & 0.2 & 0.13360957 \\ 0.05962834 & 0.18449794 & 0.05962834 & 0.13360957 & 0.2 \end{bmatrix}.$$

The segment activity coefficients computed with S2 up to eight decimal places read

$$\Gamma = (1.15654716, 0.58065158, 1.52646100, 0.91448205, 1.46544753).$$

Noting that these are the segment activity coefficients and not the chemical compound activity coefficients. So these results should not be directly compared to the compound segment activity coefficients reported by [16, 23].

Example 2 is a mixture with $n = 4$ segments, originally from [24] and also investigated by [16]. The case is actually a binary mixture. The first chemical compound can form hydrogen bonds and consists of 3 segments. The second compound has only one segment and is assumed to be *inert* (zero interaction energies with all other segment types). For the case where the inert compound is very diluted, $\theta_4 = 0.001$, the corresponding matrix reads:

$$A = \begin{bmatrix} 0.666 & 0.1665 & 0.1665 & 0.001 \\ 0.666 & 0.0536060522 & 10.7392655 & 0.001 \\ 0.666 & 10.7392655 & 0.0536060522 & 0.001 \\ 0.666 & 0.1665 & 0.1665 & 0.001 \end{bmatrix}.$$

The segment activity coefficients computed with AVS up to ten decimal places are

$$\Gamma = (1.1587320975, 0.2706845215, 0.2706845215, 1.1587320975).$$

Example 3 presents a more challenging case: a highly diluted mixture of n-octane (mole fraction 0.001) in water. The corresponding matrix A has order $n = 27$, with 5 segments from the first compound and 22 from the second. In this case, the interaction energies and segment areas were computed by the JCOSMO package [25–27]. The resulting matrix is available as supplemental online material alongside to this article. The segment activity coefficients computed with S2 up to four decimal places are

$$\begin{aligned} \Gamma = (&1.3984, 1.5461, 1.5735, 1.4678, 1.2576, 0.4989, 0.6946, 0.9271, 1.1748, \\ &1.3984, 1.5461, 1.5735, 1.4678, 1.2576, \\ &0.1153, 0.2117, 0.3680, 0.5969, 0.8909, 1.1748, 1.3984, 1.2576, \\ &0.9978, 0.7021, 0.4281, 0.2406, 0.1271). \end{aligned}$$

Residuals $r := x(Ax) - \mathbf{1}$ and iteration numbers are computed for each example and each algorithm. The results are shown in Table 1.

Table 1. *DAD* algorithms applied to Examples 1–3.

	Example 1		Example 2		Example 3	
algorithm	residual	iterations	residual	iterations	residual	iterations
S1	2.88e−14	14	8.24e−13	38	4.17e−13	87
S2	2.88e−14	14	1.75e−12	37	4.18e−13	87
JR	1.25e−12	58	2.19e−13	16	1.85e−12	334
AVS	9.24e−13	59	7.53e−14	18	1.80e−12	337
Y	3.33e−16	7	2.22e−16	7	4.44e−16	8

For all examples, Y gives the best results. This is not surprising as *Y* has quadratic convergence order while the other methods merely have linear convergence order. Comparing the linear methods, we see that for Example 1 Sinkhorn's algorithms converge faster than JR and AVS. However, for Example 2 the picture is almost reversed. For the challenging Example 3, all linear methods converge terribly slowly. Such a defect is well-known for the Sinkhorn-Knopp iteration for doubly stochastic D_1AD_2 balancing, see [28]. The convergence rate of the Sinkhorn-Knopp algorithm is linked to spectral properties of D_1AD_2, namely to its subdominant eigenvalue, see [10]. It seems plausible that convergence rates of S1, S2, JR, and AVS are likewise linked to spectral properties of the row-stochastic *DAD* scaling. However, a detailed analysis of that is beyond the scope of this article.

6. Conclusions

We have given the link between Sinkhorn's *DAD* theorem and the self-consistency equation in pairwise interaction surface models, like COSMO-RS and COSMA-SAC, widely used in physical chemistry and chemical engineering applications, see Section 1.

We have presented and implemented known iterative *DAD* algorithms which can be used for solving the self-consistency equation. Namely, these are Sinkhorn's iterations S1 and S2, the algorithm JR of Johnson and Reams, the averaged successive substitution method AVS introduced by Klamt, and a Newton-type method recently proposed by Yan, see Section 3.

We have stated a simple recursion formula for JR, see (9), which shows its similarity to AVS, see (10). The only difference between both is that JR uses the geometric mean for averaging while AVS uses the arithmetic mean.

We have proven convergence of AVS by using a classical fixed-point theorem of Edelstein for contractive mappings, see Theorem 4.2 and Corollary 4.1. In particular, this provides a new constructive proof of Sinkhorn's *DAD* theorem. Our proof is given in a general form which uniformly also shows convergence of JR and Yan's damped iteration in step (4) of his algorithm, see Listing 5. Moreover, our proof includes nonnegative matrices with positive diagonal. Nevertheless, it remains an open question whether our techniques can be extended to even wider classes of nonnegative matrices as considered in [5, Thm. 8.2].

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