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CODA IN THREE-WAY ARRAYS AND RELATIVE SAMPLE SPACES

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***Abstract**: The object of these short notes is to give a set of convenient symbols to define the sample space for the different compositional vectors that can be arranged into a three-way array. For the exploratory analysis of three-way data, Parafac/Candecomp and Tucker3 are some of the most applied models for low-rank decomposition of three-way arrays. Here, in addition to the relative geometry, is presented a concise overview as to how the elements of a three-way array can be transformed into compositional form and the relative geometry is given.*

***Keywords**: Simplex space, perturbation operation, powering, compositional data, three-mode analysis.*

1. Introduction

Compositional data (CoDa) consist of vectors of positive values summing to a unit, or in general, to some fixed constant for all vectors. They appear as proportions, percentages, concentrations, absolute and relative frequencies. Geometrically speaking, the simplex is the sample space for a compositional vector due to the nature of compositional data. The simplex has been studied as a Euclidean linear vector space [14] [5]. In these papers, most of the elements that were introduced by Aitchison in the 1980s [1] [3], such as perturbation and powering or orthogonal log-contrasts, had been organized into a systematic and coherent mathematical scheme. Recently, there have been additional developments of new tools for the representation of compositions and their exploratory analysis of the geometry of the simplex [6] [7] [8] [9]. Moreover, some important tools for developing mathematical and statistical models, consisting of the initial concepts of limit, convergence, derivates and integrals involving functions defined on the simplex are given by Egozcue et al. [10] [4].

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Through Gallo's papers [11] [12] it has been shown how three-mode analyses like the Parafac/Candecomp and Tucker3, can be used to obtain latent variables by compositional data, and that both methods yield a low-dimensional representation of the results. Like the Principal Component Analysis on two-way matrices of compositional data [2], a low-dimensional description of the three-way arrays has proved difficult to handle statistically because of the awkward constraint where the components of each vector must sum to unity. Undoubtedly, in order to examine the problems that potentially occur when a three-mode analysis on compositional data is performed, it is very important to define the sample space of these data.
The aim of these short notes is to give a set of convenient symbols to define the sample space for the different compositional vectors that can be arranged into a three-way array. Thus, the sample space can be identified through a one-to-one correspondence with the possible outcomes of the observational or experimental process.

2. Elements of simplicial geometry

2.1 Three-way array definitions

Let $\underline{\tilde{\mathbf{V}}}$ $(I \text{ x } J \text{ x } K)$ be a three-way array with *I* objects, *J* variables, and *K* occasions and generic element $\tilde{v}_{ijk} > 0 \;\; \forall\, i,j,k$. There are three types of slices referred to as $(I \text{ x } J)$ frontal $\tilde{\mathbf{V}}_k$ with $k = 1,...,K$, $(I \text{ x } K)$ vertical $\tilde{\mathbf{V}}_j$ with $j = 1,...,J$, and $(K \text{ x } J)$ horizontal $\tilde{\mathbf{V}}_i$ with $i = 1,...,I$. These slices can be concatenated between them obtaining the following matricizing of the three-way array as $\tilde{\mathbf{V}}_A$ $(I \text{ x } JK)$, $\tilde{\mathbf{V}}_B$ $(J \text{ x } IK)$, and $\tilde{\mathbf{V}}_C$ $(K \text{ x } IJ)$, i.e. $\tilde{\mathbf{V}}_A = \left[\tilde{\mathbf{V}}_1 | \ldots | \tilde{\mathbf{V}}_k | \ldots | \tilde{\mathbf{V}}_K\right]$, $\tilde{\mathbf{V}}_B = \left[\tilde{\mathbf{V}}_1^t | \ldots | \tilde{\mathbf{V}}_k^t | \ldots | \tilde{\mathbf{V}}_K^t\right]$, and $\tilde{\mathbf{V}}_C = \left[\tilde{\mathbf{V}}_1 | \ldots | \tilde{\mathbf{V}}_i | \ldots | \tilde{\mathbf{V}}_I\right]$. In addition, the three-way array $\underline{\tilde{\mathbf{V}}}$ can be broken up into vectors, called fibers. The different types of fibers are referred to as rows, columns and tubes. Thus, $\underline{\tilde{\mathbf{V}}}$ can be broken up into *IK* rows $\tilde{\mathbf{v}}_{ik}$, *JK* columns $\tilde{\mathbf{v}}_{jk}$, *IJ* tubes $\tilde{\mathbf{v}}_{ij}$, with dimension $(1 \text{ x } J)$, $(1 \text{ x } I)$ and $(1 \text{ x } K)$, respectively. Each slice of a three-way array can be converted into a column vector by vec-operator [13] [15]. Thus, it is possible to arrange all the column vectors of a matrix underneath each other, for example let $\tilde{\mathbf{V}}_i$ be the *i*th horizontal slice the $vec\left(\tilde{\mathbf{V}}_i^t\right) = \underline{\tilde{\mathbf{v}}}_i = \left[\tilde{\mathbf{v}}_{i1} | \ldots | \tilde{\mathbf{v}}_{ik} | \ldots | \tilde{\mathbf{v}}_{iK}\right]$, so it is the *i*th row of $\tilde{\mathbf{V}}_A$.

2.2 Basics concept

Let S_k^J be the simplex space with dimension $J-1$, defined as $S_k^J = \left\{\mathbf{v}_{\bullet k} = \left(v_{\bullet 1k}, \ldots, v_{\bullet Jk}\right) : \; v_{\bullet 1k} > 0, \ldots, v_{\bullet Jk} > 0; \sum_j v_{\bullet jk} = \kappa\right\}$,
where κ is a given positive constant, which is usually 1 or 100, depending on whether the variables are measured in part per unit or as percentages, respectively. A simplex element, $\mathbf{v}_{\bullet k} \in S_k^J$, is called composition, and its components $v_{\bullet jk}$ ($j = 1, \ldots, J$) are called parts of $\mathbf{v}_{\bullet k}$.

Definition 1: Let $\tilde{\mathbf{v}}_{\bullet k}$ be a row of the frontal slice $\tilde{\mathbf{V}}_k$, its closure is defined as $\mathbf{v}_{\bullet k} = \mathbb{C}\left(\tilde{\mathbf{v}}_{\bullet k}\right) = \left(\kappa \tilde{v}_{\bullet 1k} / \left\|\tilde{\mathbf{v}}_{\bullet k}\right\|, \ldots, \kappa \tilde{v}_{\bullet Jk} / \left\|\tilde{\mathbf{v}}_{\bullet k}\right\|\right) = \left(\kappa \tilde{v}_{\bullet 1k} / \sum_j \tilde{v}_{\bullet jk}, \ldots, \kappa \tilde{v}_{\bullet Jk} / \sum_j \tilde{v}_{\bullet jk}\right)$ where $\|\,.\,\|$ is the norm of vector. The operator $\mathbb{C}$ is called the κ-closure operator.

All the fibers of a three-way array $\underline{\tilde{\mathbf{V}}}$ can be transformed into compositions by the closure operator $\mathbb{C}$, but it would really only make sense to do this for the objects. Thus, it is applied to a row of $\tilde{\mathbf{V}}_k$ $\left(k = 1, \ldots, K\right)$, it then defines a transformation $\Re_+^J \to S_k^J$, where S_k^J is previously defined.

Afterwards, we proceed to indicate the *IK* rows of three-way array as compositions. Thus, for each frontal slice $\mathbf{V}_k$ $\left(k = 1, \ldots, K\right)$ we have *I* compositions with the relative sample space S_k^J $\left(k = 1, \ldots, K\right)$. Therefore, in each simplex S_k^J there are *I* points with coordinates $\mathbf{v}_{1k} \ldots \mathbf{v}_{ik} \ldots \mathbf{v}_{Ik}$.

In accord to the two basic operations, denoted by $\oplus$ and $\odot$, namely perturbation and power transformation or powering, it is possible to introduce the following basic operations [3].

Definition 2: Let $\mathbf{v}_{ik}$, $\mathbf{v}_{i'k}$ be compositions in S_k^J. Perturbation, denoted as $\oplus$, is defined as $\mathbf{v}_{ik} \oplus \mathbf{v}_{i'k} = \mathbb{C}\left(\tilde{v}_{i1k}\tilde{v}_{i'1k}, \ldots, \tilde{v}_{iJk}\tilde{v}_{i'Jk}\right)$.

Definition 3: Let $\mathbf{v}_{ik}$ be composition in S_k^J and $\alpha \in \Re$. Powering, denoted as $\odot$, is defined as $\alpha \odot \mathbf{v}_{ik} = \mathbb{C}\left(\tilde{v}_{i1k}^{\alpha}, \ldots, \tilde{v}_{iJk}^{\alpha}\right)$.

With the perturbation operation and the power transformation, the simplex S_k^J is a vector space with dimension $J - 1$ on $\Re$. The following properties make them analogous to translation and scalar multiplication.

Property 1: Let $\mathbf{v}_{ik}$, $\mathbf{v}_{i'k}$, $\mathbf{v}_{i''k}$ be compositions in S_k^J and α a real constant. Then

- (associative) $\left(\mathbf{v}_{ik} \oplus \mathbf{v}_{i'k}\right) \oplus \mathbf{v}_{i''k} = \mathbf{v}_{ik} \oplus \left(\mathbf{v}_{i'k} \oplus \mathbf{v}_{i''k}\right)$;
- (commutative) $\mathbf{v}_{ik} \oplus \mathbf{v}_{i'k} = \mathbf{v}_{i'k} \oplus \mathbf{v}_{ik}$;
- (opposite element) $\mathbf{v}_{ik} \oplus \left(-1 \odot \mathbf{v}_{i'k}\right) = \mathbf{v}_{ik} \,!\, \mathbf{v}_{ik} = \eta$;
- (neutral element) $\eta = \mathbb{C}\left(1, \ldots, 1\right) = \left(1/J, \ldots, 1/J\right)$;
- (distributive) $\left(\alpha \odot \mathbf{v}_{ik}\right) \oplus \left(\alpha \odot \mathbf{v}_{i'k}\right) = \alpha \odot \left(\mathbf{v}_{ik} \oplus \mathbf{v}_{i'k}\right)$;
- (unit) $\left(1 \odot \mathbf{v}_{ik}\right) = \mathbf{v}_{ik}$.

Note that we handle the operations $\oplus$, ! and $\odot$ in the simplex formally like we do with the standard vector operations +, - and x in multidimensional real space.

2.3 Simplex spaces for objects observed across the occasions

In three-way arrays, it is also noticed that the same object is observed on several occasions, for example the data of *i*th object are arranged in the horizontal slice $\tilde{\mathbf{V}}_i$, and often the values that

the object assumes on the different occasions are plotted in the same space as trajectory. In this case, for each object K points are plotted and linked between them in the same real space $\Re_+^J$. In the same way, if the rows of a horizontal slice are compositions its sample space can be defined as S^J, and for the representation of each composition K points are linked between them to have the trajectory of each composition through the K occasions.
On the other hand, often each object observed on several occasions is often summarized in a real space with only one point. In this case, by vec-operator, $vec(\ .\ )$, the horizontal slices can be vectored. This operator can be applied to the horizontal slices before or after the closure operator. Thus, it is possible to define the following two vectors:

$$\underline{\mathbf{v}}_i = \mathbb{C}\left(vec\left(\tilde{\mathbf{V}}_i^t\right)\right) = \left(\kappa\tilde{v}_{i11} / \|\tilde{\underline{\mathbf{v}}}_i\|,\ldots,\kappa\tilde{v}_{iJ1} / \|\tilde{\underline{\mathbf{v}}}_i\| |\ldots| \kappa\tilde{v}_{i1K} / \|\tilde{\underline{\mathbf{v}}}_i\|,\ldots,\kappa\tilde{v}_{iJK} / \|\tilde{\underline{\mathbf{v}}}_i\|\right) = \left[\mathbf{v}_{i1}|\ldots|\mathbf{v}_{ik}|\ldots|\mathbf{v}_{iK}\right]$$

$$\dot{\underline{\mathbf{v}}}_i = vec\left(\mathbb{C}\left(\tilde{\mathbf{V}}_i^t\right)\right) = \left(\kappa\tilde{v}_{i11} / \|\tilde{\mathbf{v}}_{i1}\|,\ldots,\kappa\tilde{v}_{iJ1} / \|\tilde{\mathbf{v}}_{i1}\| |\ldots| \kappa\tilde{v}_{i1K} / \|\tilde{\mathbf{v}}_{iK}\|,\ldots,\kappa\tilde{v}_{iJK} / \|\tilde{\mathbf{v}}_{iK}\|\right) = \left[\dot{\mathbf{v}}_{i1}|\ldots|\dot{\mathbf{v}}_{ik}|\ldots|\dot{\mathbf{v}}_{iK}\right]$$

The $\mathbb{C}\left(vec(\ .\ )\right)$ defines a transformation $\Re_+^{JK} \to S^{JK}$, with
$S^{JK} = \left\{\underline{\mathbf{v}}_{\bullet} = \left(v_{\bullet 11},\ldots,v_{\bullet J1}|\ldots|v_{\bullet 1K},\ldots,v_{\bullet JK}\right):\ v_{\bullet jk} > 0 \text{ for all elements}; \sum_j \sum_k v_{\bullet jk} = \kappa\right\}$.
By contrast, the $vec\left(\mathbb{C}(\ .\ )\right)$ defines before a transformation of each single vector $\tilde{\mathbf{v}}_{\bullet k}$ a transformation in the simplex space ${}^k\Re_+^J \to S_k^J$, then by vec-operator we have the simplex space $S_1^J \text{ x} \ldots \text{x } S_k^J \text{ x} \ldots \text{x } S_K^J = \prod_{k=1}^{K} S_k^J = \dot{S}^{JK}$.
In accordance with the Definitions 2 and 3, it is possible to verify the Property 1 for the ternary $\left(S^{JK},\oplus,\odot\right)$ and $\left(\dot{S}^{JK},\oplus,\odot\right)$. Besides, to investigate the dimension of the vector spaces S^{JK} and $\dot{S}^{JK}$ the additive log-ratio transformation (*alr*) can be used [3].

Definition 4: Let $\underline{\mathbf{v}}_i$ and $\dot{\underline{\mathbf{v}}}_i$ be a compositional vector and a vector with K juxtaposed compositional vectors, respectively. The transformation $alr\left(\underline{\mathbf{v}}_i\right):\ S^{JK} \to \Re^{JK-1}$; is defined as $alr\left(\underline{\mathbf{v}}_i\right) = \log\left(\frac{v_{i11}}{v_{iJK}},\ldots,\frac{v_{iJ1}}{v_{iJK}}|\ldots|\frac{v_{i1k}}{v_{iJK}},\ldots,\frac{v_{iJk}}{v_{iJK}}|\ldots|\frac{v_{i1K}}{v_{iJK}},\ldots,\frac{v_{i(J-1)K}}{v_{iJK}}\right)$.
In a similar way, the transformation $alr\left(\dot{\underline{\mathbf{v}}}_i\right):\ \dot{S}^{JK} \to \Re^{K(J-1)}$; is defined as $alr\left(\dot{\underline{\mathbf{v}}}_i\right) = \log\left(\frac{v_{i11}}{v_{iJ1}},\ldots,\frac{v_{i(J-1)1}}{v_{iJ1}}|\ldots|\frac{v_{i1k}}{v_{iJk}},\ldots,\frac{v_{i(J-1)k}}{v_{iJk}}|\ldots|\frac{v_{i1K}}{v_{iJK}},\ldots,\frac{v_{i(J-1)K}}{v_{iJK}}\right)$.

Proposition 1: Let be $\underline{\mathbf{v}}_i^* \in \Re^{JK-1}$, the transformation $alr\left(\underline{\mathbf{v}}_i\right):\ S^{JK} \to \Re^{JK-1}$ is one-to-one if the inverse additive log-ratio transformation $alr^{-1}\left(\underline{\mathbf{v}}_i\right)$ is
$alr^{-1}\left(\underline{\mathbf{v}}_i^*\right) = \mathbb{C}\left[\exp\left(v_{i11}^*,\ldots,v_{iJ1}^*|\ldots|v_{i1k}^*,\ldots,v_{iJk}^*|\ldots|v_{i1K}^*,\ldots,v_{i(J-1)k}^*,0\right)\right]$.

In a similar way it is possible to introduce the additive log-ratio for $\underline{\dot{\mathbf{v}}}_i$.

Proposition 2: Let be $\underline{\dot{\mathbf{v}}}_i^* \in \Re^{K(J-1)}$, the transformation $a\hat{l}r\left(\underline{\dot{\mathbf{v}}}_i\right)$: $\dot{S}^{JK} \to \Re^{K(J-1)}$ is one-to-one if the inverse additive log-ratio transformation $a\hat{l}r^{-1}\left(\underline{\dot{\mathbf{v}}}_i\right)$ is:

$$a\hat{l}r^{-1}\left(\underline{\dot{\mathbf{v}}}_i^*\right) = \left\{\mathbb{C}\left[\exp\left(v_{i11}^*,\ldots,v_{i(J-1)1}^*,0\right)\right]\middle|\ldots\middle|\mathbb{C}\left[\exp\left(v_{i11}^*,\ldots,v_{i(J-1)1}^*,0\right)\right]\middle|\ldots\middle|\mathbb{C}\left[\exp\left(v_{i11}^*,\ldots,v_{i(J-1)1}^*\right)\right]\right\}.$$

Both alr and $a\hat{l}r$ transformations are isomorphism of vector spaces.

Proof. The $alr^{-1}\left(alr\left(\underline{\mathbf{v}}_i\right)\right) = \underline{\mathbf{v}}_i$, then for any composition in the simplex space S^{JK}, the additive log-ratio is a one-to-one transformation in the real space $\Re^{JK-1}$. On the other hand, the isomorphism requires that, for all $\underline{\mathbf{v}}_i, \underline{\mathbf{v}}_{i'} \in S^{JK}$, and $\alpha, \beta \in \Re$,

$alr\left[\left(\alpha \odot \underline{\mathbf{v}}_i\right) \oplus \left(\beta \odot \underline{\mathbf{v}}_{i'}\right)\right] = \alpha \cdot alr\left(\underline{\mathbf{v}}_i\right) + \beta \cdot alr\left(\underline{\mathbf{v}}_{i'}\right)$

which holds from Definitions 2 and 3.

These results can be easy to verify as to $a\hat{l}r\left(\underline{\dot{\mathbf{v}}}_i\right)$ too. In fact, the $alr^{-1}\left(alr\left(\underline{\dot{\mathbf{v}}}_i\right)\right) = \underline{\dot{\mathbf{v}}}_i$ then by the $a\hat{l}r$ transformation in the simplex space $\dot{S}^{JK}$ are transformed into one point in the real space $\Re^{K(J-1)}$. Besides, the isomorphism for all $\underline{\dot{\mathbf{v}}}_i, \underline{\dot{\mathbf{v}}}_{i'} \in \dot{S}^{JK}$, let be $\alpha, \beta \in \Re$, requires that

$a\hat{l}r\left[\left(\alpha \odot \underline{\dot{\mathbf{v}}}_i\right) \oplus \left(\beta \odot \underline{\dot{\mathbf{v}}}_{i'}\right)\right] = \alpha \cdot a\hat{l}r\left(\underline{\dot{\mathbf{v}}}_i\right) + \beta \cdot a\hat{l}r\left(\underline{\dot{\mathbf{v}}}_{i'}\right)$

which holds again from Definitions 2 and 3.

Definition 5: Given a compositional vector $\underline{\mathbf{v}}_{\bullet} \in S^{JK}$ a subcomposition $\mathbf{v}_{\bullet k}^s$ with J parts is obtained applying the closure operation to a subvector $\mathbf{v}_{\bullet k}$ of $\underline{\mathbf{v}}_{\bullet}$: $\mathbf{v}_{\bullet k}^s = \mathbb{C}\left(\mathbf{v}_{\bullet k}\right)$.

In compositional analysis an important feature is that the ratio of any two components of a subcomposition is the same as the ratio of the corresponding two components in the full composition.

It is possible to show that each subvector $\dot{\mathbf{v}}_{\bullet k}$ of $\underline{\dot{\mathbf{v}}}_{\bullet}$ is equal to the vector of the juxtaposed subcompositions $\mathbf{v}_{\bullet k}^s$ given by applying the closure operation to the subvector $\mathbf{v}_{\bullet k}$ of $\underline{\mathbf{v}}_{\bullet}$: $\underline{\dot{\mathbf{v}}}_{\bullet} = \left[\dot{\mathbf{v}}_{\bullet 1}\middle|\ldots\middle|\dot{\mathbf{v}}_{\bullet K}\right] = \left[\mathbb{C}\left(\mathbf{v}_{\bullet 1}\right)\middle|\ldots\middle|\mathbb{C}\left(\mathbf{v}_{\bullet K}\right)\right]$.

From a geometrical point of view, it is possible to observe that each subcomposition $\mathbf{v}_{\bullet k}$ of $\underline{\mathbf{v}}_{\bullet}$ is a projection of $\underline{\mathbf{v}}_{\bullet}$ in S_k^J. In other words, a subcomposition can be regarded as a composition in a simplex of lower dimension than that of the full composition. Thus, the simplex spaces S_k^J $\left(k = 1,\ldots,K\right)$ are the subsimplexes of S^{JK}.

References

[1]. Aitchison, J. (1982). The statistical analysis of compositional data (with discussion). *Journal of the Royal Statistical Society, Series B (Methodological)*, 44(2), 139-177.

[2]. Aitchison, J. (1983). Principal component analysis of compositional data, *Biometrika*, 70 (1), 57–65.

[3]. Aitchison, J. (1986). *The statistical analysis of compositional data*, Monographs on Statistics and Applied Probability. Chapman & Hall, Ltd, London, UK.

[4]. Barceló-Vidal, C., Martín-Fernández, J.A., Mateu-Figueras, G. (2011). Compositional differential calculus on the simplex, in *Compositional Data Analysis: Theory and Applications*, eds. V. Pawlowsky-Glahn and A. Buccianti, John Wiley & Sons, Ltd, Chichester, UK.

[5]. Billheimer, D., Guttorp, P., Fagan, W. (2001), Statistical interpretation of species composition, *Journal of American Statistics Association*, 96 (456), 1205–1214.

[6]. Egozcue, J.J., Pawlowsky-Glahn, V. (2005). Groups of parts and their balances in compositional data analysis, *Mathematical Geology*, 37 (7), 795–828.

[7]. Egozcue, J.J., Pawlowsky-Glahn, V. (2006). Exploring compositional data with the coda-dendrogram, in IAMG 2006: *The XIth annual conference of the International Association for Mathematical Geology*, Liege - Belgium, 3-8 September 2006.

[8]. Egozcue, J.J., Pawlowsky-Glahn, V., Mateu-Figueras, G., Barcelo-Vidal, C. (2003). Isometric logratio transformations for compositional data analysis, *Mathematical Geology*, 35 (3), 279-300.

[9]. Egozcue, J.J., Barceló-Vidal, C., Martín-Fernández, J.A., Jarauta-Bragulat, E., Díaz-Barrero, J.L., Mateu-Figueras, G. (2011). Elements of Simplicial Linear Algebra and Geometry, in *Compositional Data Analysis: Theory and Applications*, eds. V. Pawlowsky-Glahn and A. Buccianti, John Wiley & Sons, Ltd, Chichester, UK.

[10]. Egozcue, J.J., Jarauta-Bragulat, E., Díaz-Barrero, J.L. (2011). Calculus of simplex-valued functions, in *Compositional Data Analysis: Theory and Applications*, eds. V. Pawlowsky-Glahn and A. Buccianti, John Wiley & Sons, Ltd, Chichester, UK.

[11]. Gallo, M. (2013). Log-ratio and parallel factor analysis: an approach to analyze three-way compositional data, in *Advances in intelligent and soft computing special issue Dynamics of socio-economic systems*. Springer. In Press. [11]

[12]. Gallo, M. (2012). *Tucker3 analysis of compositional data*. Submitted. [12]

[13]. Kiers, H. A. L. (2000). Towards a standardized notation and terminology in multiway analysis. *Journal of Chemometrics*, 14, 105–122.

[14]. Pawlowsky-Glahn, V., Egozcue, J.J. (2001). Geometric approach to statistical analysis on the simplex. *Stochastic Environmental Research and Risk Assessment*, 15 (5), 384-398.

[15]. Smilde, K.A., Bro, R., Geladi, P. (2004). *Multi-way analysis: applications in the chemical sciences*, Wiley, Chichester, UK.