Talanta 148 (2016) 144–152

Contents lists available at ScienceDirect

Talanta

journal homepage: www.elsevier.com/locate/talanta

Three-way analysis of the UPLC–PDA dataset for the multicomponent quantitation of hydrochlorothiazide and olmesartan medoxomil in tablets by parallel factor analysis and three-way partial least squares

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ARTICLE INFO

Article history:
Received 14 July 2015
Received in revised form
14 October 2015
Accepted 24 October 2015
Available online 26 October 2015

Keywords:
Parallel factor analysis
Three-way partial least squares
Chromatographic spectral three-way array
Hydrochlorothiazide
Olmesartan medoxomil
Tablet

ABSTRACT

An application of parallel factor analysis (PARAFAC) and three-way partial least squares (3W-PLS1) regression models to ultra-performance liquid chromatography–photodiode array detection (UPLC–PDA) data with co-eluted peaks in the same wavelength and time regions was described for the multicomponent quantitation of hydrochlorothiazide (HCT) and olmesartan medoxomil (OLM) in tablets. Three-way dataset of HCT and OLM in their binary mixtures containing telmisartan (IS) as an internal standard was recorded with a UPLC–PDA instrument. Firstly, the PARAFAC algorithm was applied for the decomposition of three-way UPLC–PDA data into the chromatographic, spectral and concentration profiles to quantify the concerned compounds. Secondly, 3W-PLS1 approach was subjected to the decomposition of a tensor consisting of three-way UPLC–PDA data into a set of triads to build 3W-PLS1 regression for the analysis of the same compounds in samples. For the proposed three-way analysis methods in the regression and prediction steps, the applicability and validity of PARAFAC and 3W-PLS1 models were checked by analyzing the synthetic mixture samples, inter-day and intra-day samples, and standard addition samples containing HCT and OLM. Two different three-way analysis methods, PARAFAC and 3W-PLS1, were successfully applied to the quantitative estimation of the solid dosage form containing HCT and OLM. Regression and prediction results provided from three-way analysis were compared with those obtained by traditional UPLC method.

1. Introduction

Hydrochlorothiazide (HCT), is a diuretic agent commonly used in combination with other antihypertensive drugs. HCT increases the excretion of chloride, sodium and potassium along with water therefore reduces plasma volume and decreases the blood pressure [1]. Olmesartan medoxomil (OLM) is an antihypertensive drug within the group angiotensin II receptor blockers. Its active metabolite, olmesartan, reduces blood pressure by binding to AT1 receptors in vascular muscle and inhibiting the physiological effect of angiotensin II [2]. Studies have shown that numerous hypertensive patients need more than one antihypertensive drugs to keep blood pressure under control. Diuretic effect of HCT and antihypertensive effect of OLM have harmonious impacts on cardiovascular system. Hence, combination of HCT and OLM is a useful and common way in hypertension treatment [3–5].

There have been several methods to quantify HCT and OLM in their binary mixture in pharmaceutical preparations by UV-spectrophotometry [6–8], HPLC [9–12], and HPTLC [12,13] and in biological fluids by LC–MS/MS [14,15].

High-performance liquid chromatography with photodiode array detection (HPLC–PDA) is the most common method applied for the qualitative and quantitative analyses of raw material, environmental organic pollutants, drug substance, drug preparation and active compounds in biological fluids.

In recent years, ultra-performance liquid chromatography (UPLC) method produced by Waters offers a relatively new approach providing new opportunities in liquid chromatography and its applications, particularly regarding decrease of the run time and solvent consumption. Due to the mentioned advantages, UPLC technique instead of HPLC has been preferred for the chemical and pharmaceutical analysis. UPLC–PDA technique allows getting two-way UPLC–PDA data by measuring absorbance (A) as a function of time (t) and wavelength (λ), defined by the equation $A=f(t,\lambda)$, for each sample of analytes.

In all cases, UPLC method as well as conventional HPLC method may give inadequate elution of the peaks (or overlapping peaks) in

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http://dx.doi.org/10.1016/j.talanta.2015.10.074
0039-9140/

a chromatogram due to similar physical and chemical properties of analytes. This problem can be resolved using two operational approaches either additional chromatographic experiments and studies or chemometrics.

Taking into account the above overlapping chromatographic peaks of substances, three-way data analysis methods e.g. parallel factor analysis (PARAFAC) [16], Tucker3 [17], and three-way partial least squares (3W-PLS1) [19] which are popular methods, can be applied for the qualitative and quantitative estimation of multicomponent mixtures without using additional separation procedures with acceptable precision and accuracy. Another approach, multivariate curve resolution-alternating least squares (MCR-ALS) has also been used for the co-elution problems in chromatographic analysis [18]. In chemometrics, pretreatments namely baseline and time shift correction, centering and scaling of three-way data array have been used to increase the quality of the analysis, especially to get more precise and accurate analysis results [20,21].

In this study, PARAFAC and 3W-PLS1 were applied to the co-eluted UPLC peaks for the multicomponent quantitation of HCT and OLM in tablets using the internal standard, telmisartan. Assay results provided by applying three-way analysis methods (PARAFAC and 3W-PLS1) were compared with those obtained by newly developed traditional UPLC method based on the univariate calibration.

2. Theory

2.1. PARAFAC model

Parallel factor analysis (PARAFAC), which is also expressed as canonical decomposition (CANDECOMP), is one of popular three-way analysis methods [22,23]. The PARAFAC model is based on the decomposition of three-way data (or n-way data) into tri-linear (or multi-linear) component using different algorithms (alternating least squares (ALS) approach, in our case) [16,19,24,29]. The PARAFAC is similar to the decomposition of two-way data using the principal component analysis (PCA). When PARAFAC model is conceptually compared with PCA, the PARAFAC solution is unique under Kruskal's conditions, and it does not need to rotate the loadings as in PCA [30]. The decomposition of a three-way array $\underline{X}$ with $I \times J \times K$ dimension into three loadings matrices, which correspond to the modes/ways a (i, f), b (j, f) and c (k, f) of the related $\underline{X}$, can be written for the PARAFAC model as follows

$$x_{ijk} = \sum_{f=1}^{F} a_{if} b_{jf} c_{kf} + e_{ijk} \quad (1)$$

where x_{ijk} is an element of $\underline{X}$ with $i=1,\ldots, I$, $j=1,\ldots, J$ and $k=1,\ldots, K$; F is the number of components used in three modes of $\underline{X}$; $a_{if}b_{jf}$ and c_{kf} are the elements of the vectors $\boldsymbol{a_f}$,$\boldsymbol{b_f}$ and $\boldsymbol{c_f}$, respectively; e_{ijk} is an element of the residual data array $\underline{E}$. The PARAFAC decomposition of a three-way array $\underline{X}$ of the UPLC–PDA data for F-components is schematically illustrated in Fig. 1.

As it can be seen from Fig. 1, PARAFAC decomposition for each component gives us three loadings, which correspond to pure chromatograms (a_f), pure spectra (b_f) and relative concentration set (c_f). An iterative ALS algorithmic computation is used to fit the PARAFAC model of three-way data minimizing the sum of the squares of the residual data representing e_{ijk} in the fitting model [24].

2.2. Trilinear (3W) PLS model

Trilinear PLS (or multilinear PLS, which is also called as N-PLS) regression is an extension and generalization of the ordinary two-way PLS algorithm to three-way data array (or multiway data array). The algorithmic basis of the N-PLS was introduced by Bro [19] and elaborated by Smilde [25] and de Jong [26] and then the model was improved by Bro et al. [27]. As described in previous studies, the essential of three-way PLS (3W-PLS1) approach is a decomposition of the tensor $\underline{X}$ with dimensions ($I \times J \times K$) into a set of triads. In our case, it is considered three-way version of N-PLS. A matrix Z ($J \times K$) is obtained as the inner product of $\underline{X}$ (independent variable) and Y (dependent variable). Each element of Z can be written in the following form:

$$Z_{jk} = \sum_{i=1}^{I} y_i x_{ijk} \quad (2)$$

where y_i is the element of Y and x_{ijk} denotes an element of the concerned $\underline{X}$ with $i=1, \ldots., I$, $j=1, \ldots., J$ and $k=1, \ldots., K$. A set of weight vectors $\mathbf{w}^J$ and $\mathbf{w}^K$ is found as first left and right singular vectors from singular value decomposition of Z ($J \times K$). Trilinear PLS model for each element of the tensor $\underline{X}$ can be given as

$$x_{ijk} = \sum_{f=1}^{F} t_{if} w_{jf} w_{kf} + e_{ijk} \quad (3)$$

here W^J and W^K are the weight vectors giving a score t, calculated by the following equation $t = X(\boldsymbol{w}^K \otimes \boldsymbol{w}^J)$ using a matricized (unfolded) version X of the data cube, and e_{ijk} is the model error. The optimization criterion can be given for the three-way PLS model in the following formula.

$$\max_{w^J w^K} \left\{ \mathrm{cov}(t, y)/\min \sum_{i=1}^{I} \sum_{j=1}^{J} \sum_{k=1}^{K} (x_{ijk} - t_i w_j^J w_k^K)^2 \right\} \quad (4)$$

As in two-way PLS models, the algorithms of the three-way PLS models are expressed as tri-PLS1 (3W-PLS1) and tri-PLS2 (3W-PLS2) in the cases of one dependent variable and several dependent variables, respectively. The more details of trilinear PLS (or N-PLS) regression and its applications can be found in the literature [20].

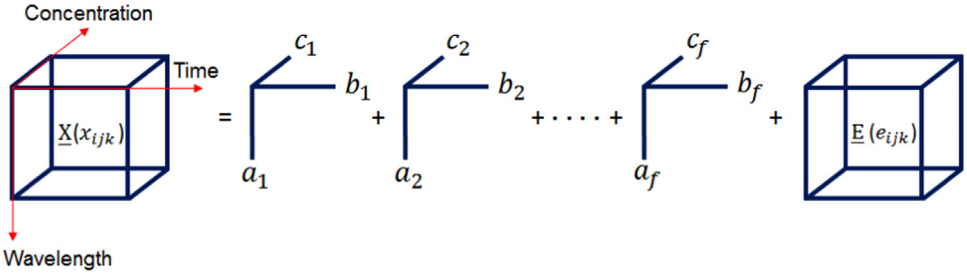

Fig. 1. Schematic illustration of the decomposition of three-way data array using the PARAFAC algorithm.