

# Chapter 2

## Mathematical and statistical background

This Chapter overviews the mathematical and statistical techniques that are adopted in the development of the multivariate and multiresolution quality monitoring strategies.

Details about the multivariate statistical techniques and the multiresolution wavelet transformation are presented and discussed. In particular, the theoretical formulation of PCA and PLS is recalled. After that, it is shown how these techniques can be integrated in monitoring frameworks of batch processes. Finally, the wavelet transform techniques are reviewed, describing their ability to extract the properties of a signal through a multiscale decomposition.

### 2.1 Multivariate statistical techniques

In the following sections, the mathematical and statistical background of the multivariate statistical techniques used in this Thesis is overviewed. In particular, details are given on both the principal component analysis and the projection on latent structures, from both the theoretical and the algorithmic points of view.

#### 2.1.1 Principal component analysis (PCA)

PCA is a multivariate statistical method that allows to summarize the information of a wide set of correlated data projecting them onto few fictitious orthogonal variables which capture the variability of and the correlation between the original data.

Let suppose that a set of data (i.e.:  $I$  observations of  $J$  variables) are collected in an  $(I \times J)$   $\mathbf{X}$  matrix from an in-control reference, after being conveniently pre-treated (see §2.1.1.2). PCA performs a decomposition of the original variables to a system of eigenvalues of the covariance matrix of  $\mathbf{X}$  through a principal axis transformation (Jackson, 1991). In this way, PCA can find the combination of the  $J$  original variables that describe the most meaningful trend of the dataset. From a mathematical point of view, PCA relies on an eigenvector decomposition of the covariance matrix of  $\mathbf{X}$ :

$$\Sigma = \text{cov}(\mathbf{X}) \quad . \quad (2.1)$$

This method splits the  $\mathbf{X}$  matrix of rank  $R$  in a sum of  $R$  matrices  $\mathbf{M}_r$  of rank 1:

$$\mathbf{X} = \mathbf{M}_1 + \mathbf{M}_2 + \dots + \mathbf{M}_r + \dots + \mathbf{M}_R \quad , \quad (2.2)$$

in which every  $\mathbf{M}_r$  matrix can be represented by the outer product of two vectors: the scores  $\mathbf{t}_r$  and the loadings  $\mathbf{p}_r$ :

$$\mathbf{X} = \mathbf{t}_1 \mathbf{p}_1^T + \mathbf{t}_2 \mathbf{p}_2^T + \dots + \mathbf{t}_r \mathbf{p}_r^T + \dots + \mathbf{t}_R \mathbf{p}_R^T \quad , \quad (2.3)$$

where  $\mathbf{p}_r^T$  is the transpose of  $\mathbf{p}_r$ . This operation is a principal axis transformation that shifts the data in a set of uncorrelated data  $\mathbf{t}_r$  described by orthogonal loading vectors  $\mathbf{p}_r$ .

In fact, the simplest way to reduce the dimensionality of the original dataset is to find a standardized linear combination  $\mathbf{p}_r^T \mathbf{X}$  of the original variables (Härdle and Simar, 2007), which maximizes the covariance of the system to deal with the correlation between the original  $J$  variables:

$$\arg \max_{\{\mathbf{p}_r: \|\mathbf{p}_r\|=1\}} [\text{cov}(\mathbf{p}_r^T \mathbf{X})] = \arg \max_{\{\mathbf{p}_r: \|\mathbf{p}_r\|=1\}} [\mathbf{p}_r^T \text{cov}(\mathbf{X}) \mathbf{p}_r] = \arg \max_{\{\mathbf{p}_r: \|\mathbf{p}_r\|=1\}} (\mathbf{p}_r^T \mathbf{\Sigma} \mathbf{p}_r) \quad \text{with } r = 1, \dots, R \quad . \quad (2.4)$$

The solution of the optimization problem corresponds to the maximization of a quadratic form for points on a unit sphere, which is the following eigenvector problem (Johnson and Wichern, 2007):

$$\arg \max_{\{\mathbf{p}_r: \|\mathbf{p}_r\|=1\}} (\mathbf{p}_r^T \mathbf{\Sigma} \mathbf{p}_r) = \lambda_r \quad \text{with } r = 1, \dots, R \quad , \quad (2.5)$$

where the loadings  $\mathbf{p}_r$  are eigenvectors of  $\mathbf{\Sigma}$ , and  $\lambda_r$  are the eigenvalues associated to  $\mathbf{p}_r$ :

$$\mathbf{\Sigma} \mathbf{p}_r - \lambda_r \mathbf{I} \mathbf{p}_r = 0 \quad , \quad (2.6)$$

being  $\mathbf{I}$  the identity matrix, and  $\mathbf{p}_r$  the director cosines of the new coordination system on which the original data are projected. As a result,  $\lambda_r$  is a measure of the variance explained by the product  $\mathbf{t}_r \mathbf{p}_r^T$ , where variance assume the meaning of quantity of information embedded into the model. Geometrically, the scores are orthogonal:

$$\begin{cases} \text{var}(\mathbf{t}_r) = \mathbf{p}_r^T \mathbf{\Sigma} \mathbf{p}_r = \lambda_r \\ \text{cov}(\mathbf{t}_r, \mathbf{t}_s) = \mathbf{p}_r^T \mathbf{\Sigma} \mathbf{p}_s = 0 \quad \text{for } r \neq s \end{cases} \quad , \quad (2.7)$$

while the loadings are orthonormal:

$$\begin{cases} \mathbf{p}_r^T \mathbf{p}_s = 0 \quad \text{for } r \neq s \\ \mathbf{p}_r^T \mathbf{p}_s = 1 \quad \text{for } r = s \end{cases} \quad . \quad (2.8)$$

Furthermore, if the PCs are zero mean and zero covariance, it follows that:

$$\begin{cases} \sum_{r=1}^R \text{var}(\mathbf{t}_r) = \text{tr}(\mathbf{\Sigma}) \\ \prod_{r=1}^R \text{var}(\mathbf{t}_r) = |\mathbf{\Sigma}| \end{cases} \quad (2.9)$$

where  $\text{tr}(\mathbf{\Sigma})$  is the trace of the covariance matrix and  $|\mathbf{\Sigma}|$  is the determinant of  $\mathbf{\Sigma}$ .

As underlined by Jackson (1991), the new variables  $\mathbf{t}_r$  are principal components of  $\mathbf{X}$  and the terms of equation (2.3) are usually presented in descending order of the eigenvalues (explained variance).

When data have a large number of highly correlated variables,  $\mathbf{X}$  is not a full rank matrix and it is possible to represent it through a small number of PCs, in such a way that the greatest part of the variance can be captured by a limited number of latent variables, defining  $A$  PCs with  $A \ll J$ . In this sense, PCA summarizes the valuable information of the original  $J$ -dimensional hyperspace of the process variables by projecting the original observations onto an  $A$ -dimensional latent subspace of PCs. From an algebraic point of view,  $\mathbf{X}$  can be described by two terms: *i*) the sum of outer products of the first  $A$  pairs scores-loadings, and *ii*) the sum of the last  $(R-A-1)$  pairs of scores and loadings:

$$\mathbf{X} = \sum_{r=1}^A \mathbf{t}_r \mathbf{p}_r^T + \sum_{r=A+1}^R \mathbf{t}_r \mathbf{p}_r^T = \mathbf{TP}^T + \mathbf{E} \quad (2.10)$$

This means that  $\mathbf{X}$  can be approximated by  $\hat{\mathbf{X}}$ :

$$\hat{\mathbf{X}} = \mathbf{TP}^T \quad (2.11)$$

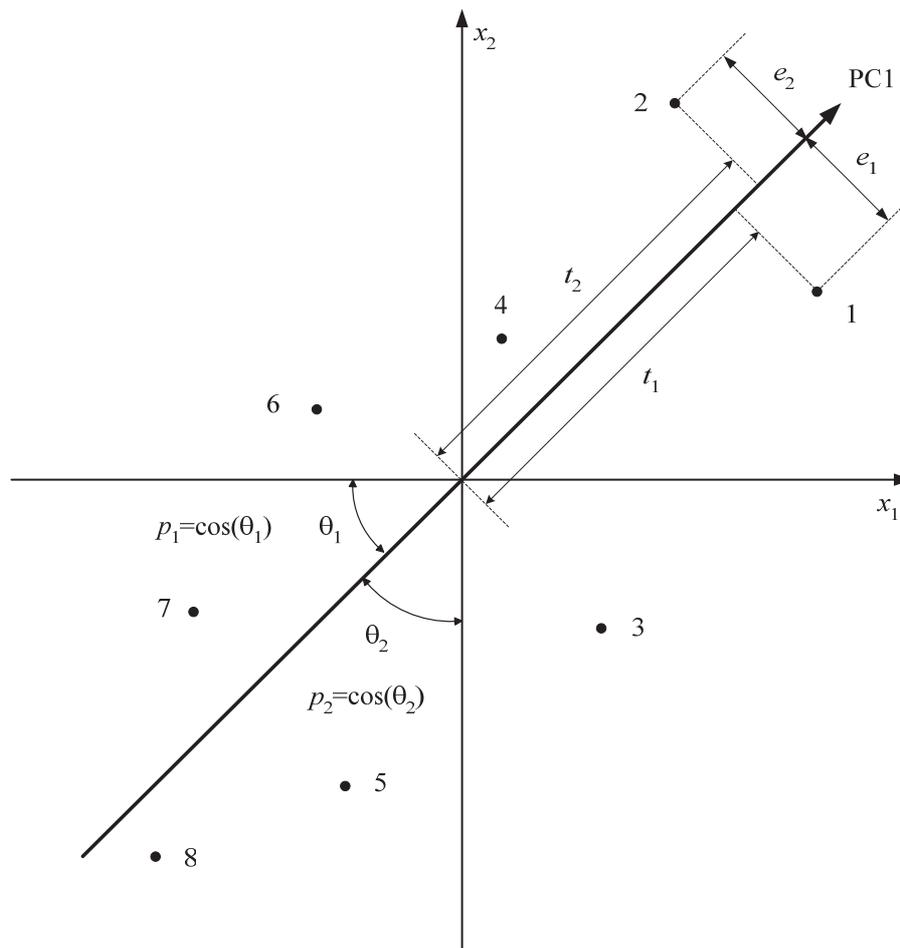
and:

$$\mathbf{E} = \mathbf{X} - \hat{\mathbf{X}} \quad (2.12)$$

is called residual and corresponds to the last  $(R-A-1)$  terms of the equation (2.3):

$$\mathbf{E} = \mathbf{t}_{A+1} \mathbf{p}_{A+1}^T + \dots + \mathbf{t}_R \mathbf{p}_R^T = \sum_{r=A+1}^R \mathbf{t}_r \mathbf{p}_r^T \quad (2.13)$$

The residuals are related to the non-systematic part of the signal (i.e. noise), that is negligible if the number of the  $A$  retained PCs is conveniently chosen.



**Figure 2.1** Geometrical interpretation of the scores and the loadings of the PCA method for a dataset with  $I=8$  observations of  $J=2$  variables ( $x_1$  and  $x_2$ ).

The geometrical interpretation of scores, loadings and residuals is shown in the simplified case of Figure 2.1. The first principal component (PC1) identifies the direction of maximum variability of the observation in the original space of process variables ( $x_1, x_2$ ). PC1 is the best fitting of the data, in the sense of a least squares minimization of the residuals.

The loadings, which are collected into matrix  $\mathbf{P}$  of dimension  $(J \times A)$ , are the director cosines of the PCs, i.e. the direction of maximum variance of the data. The scores are collected into  $\mathbf{T}$ , a  $(I \times A)$  matrix, and are the coordinates of the data into the new system of PCs. In this way it is possible to markedly reduce the number of variables, projecting the data from the high dimensional space of the original variables to a subspace of a reduced number of latent variables. The lack of accuracy of this representation is highlighted by the residual, the perpendicular distance of the data form the hyper-plane of the PCs.

### 2.1.1.1 PCA algorithm

The most frequently used algorithm for the PCA modelling is the NIPALS (non-iterative partial least squares) algorithm, a computing procedure developed by Wold, as referred by

Eriksson *et al.* (2001). This procedure computes the scores and loadings, beginning from the identification of PC1 through  $\mathbf{t}_1$  and  $\mathbf{p}_1$  for the matrix  $\mathbf{X}$ . Then, the first residual  $\mathbf{E}_1$  is calculated as the difference between  $\mathbf{t}_1\mathbf{p}_1^T$  and  $\mathbf{X}$ . In turn, the scores and loadings of  $\mathbf{E}_1$  are calculated, identifying the second principal component (PC2). The algorithm is then recursively repeated as much times as the desired number of PCs to be considered.

For an iteration  $r$ , the algorithm goes through the following steps:

1. let  $\mathbf{x}_i$  be a row vector (observation) of  $\mathbf{X}_r$ . Set:

$$\mathbf{t}_r = \mathbf{x}_i \quad ; \quad (2.14)$$

2. calculate  $\mathbf{p}_r^T$ :

$$\mathbf{p}_r^T = \frac{\mathbf{t}_r^T \mathbf{X}}{\mathbf{t}_r^T \mathbf{t}_r} \quad ; \quad (2.15)$$

3. normalize  $\mathbf{p}_r^T$  to unit length:

$$\mathbf{p}_{r,\text{new}}^T = \frac{\mathbf{p}_{r,\text{new}}^T}{\|\mathbf{p}_{r,\text{old}}^T\|} \quad ; \quad (2.16)$$

4. calculate  $\mathbf{t}_r$ :

$$\mathbf{t}_r = \frac{\mathbf{X}\mathbf{p}_r}{\mathbf{p}_r^T \mathbf{p}_r} \quad ; \quad (2.17)$$

5. compare  $\mathbf{t}_r$  calculated in 2 to the one calculated in 4. If they are equal for less than an assigned tolerance then the method is converged, else restart from 2 with the last calculated value of  $\mathbf{t}_r$ ;

6. calculate:

$$\mathbf{E}_r = \mathbf{E}_{r-1} - \mathbf{t}_r\mathbf{p}_r^T \quad (2.18)$$

7. the residual at iteration  $r$  is the matrix to begin the iteration  $(r+1)$ , setting  $\mathbf{X}_{r+1} = \mathbf{E}_r$ .

Geladi and Kowalski (1986) demonstrated that the solution of the algorithm is the same as the rigorous procedure, which solves the eigenvalue-eigenvector problem.

Modified versions, such as the straightforward implementation of modified PLS (SIMPLS, due to de Jong, 1993), and improvements of the algorithms (Yabuki and MacGregor, 1997; Lindgren and Rännar, 1998) are available in literature.

To build a data-driven model, databases of historical records have to be consulted. However, the calibration of the model presents some issues that exceeds the mere collection of the data, for example:

- pretreatment of the dataset and selection of the variables to include into the model;
- choice of the dimensionality of the subspace of PCs;
- management of the missing data in the original dataset and during the online implementations;
- treatment of the data non-linearity;
- treatment of the time-varying nature of the data in batch processes.

These issues are discussed in the following sections.

### 2.1.1.2 Data collection, variable selection and data pre-treatment

The most representative samples of in-spec products or of in-control processes have to be chosen to calibrate the model (Eriksson *et al.*, 2001; Capron *et al.*, 2005). The reference dataset should characterize the process of interest for both the target value and the admissible variability, collecting a series of historical information which form the NOC.

After properly selecting the optimal reference, the input data have to be properly pre-treated. Accordingly, some considerations should be done about: *i*) the pre-treatment of the input data, and *ii*) the importance of the variables included into the models. Researchers agree when stating that the performances of a data-driven model improves if the variables included in the model are suitably selected and conveniently pre-treated (Höskuldsson, 2001; Chu *et al.*, 2004).

Depending on the requirements, variables can be included or discarded following a criterion of engineering judgment. From the statistical point of view, the most noteworthy variables can be identified by the loadings, which express the relation between variables and the importance of the variables in PCA models. Furthermore, the variables can be weighted all in the same way, by “auto-scaling” the input data (mean-centring and scaling to unit variance) to linearize the data and to handle the differences in scales (Geladi and Kowalski, 1986; Kourti, 2003). As an alternative, different weights can be assigned to the variables to determine uneven influences of each variable on the model (Xu *et al.*, 2006).

In this Thesis, the auto-scaling is adopted. The auto-scaling mean-centres and scales to unit variance all the variables. The mean-centring is performed calculating the mean value  $\bar{x}_j$  of every variable  $j=1, \dots, J$  of the  $\mathbf{X}$  data:

$$\bar{x}_j = \frac{\sum_{i=1}^I x_{i,j}}{I}, \quad j = 1, \dots, J, \quad (2.19)$$

where  $x_{i,j}$  is the element of  $\mathbf{X}$  in row  $i$  and column  $j$ . The next step is subtracting from every variable  $\mathbf{x}_j$  (the  $j^{\text{th}}$  column vector of  $\mathbf{X}$ ) the respective mean  $\bar{\mathbf{x}}_j$ .

The scaling to unit variance is performed by dividing the values of all the variables  $\mathbf{x}_j$  in such a way that each variable gets unit variance:

$$\text{var}(\mathbf{x}_j) = \frac{\sum_{i=1}^I (x_{i,j} - \bar{\mathbf{x}}_j)^2}{I} \quad (2.20)$$

This procedure gives the same weight to all the variables and deals with the differences in the scales of the variables that have different measurement units. Furthermore, when the  $\mathbf{X}$  matrix is auto-scaled, the covariance matrix becomes the correlation matrix. For this reason PCA on auto-scaled data is considered a correlative model which extracts the correlation between variables and the loadings are the director cosines of the new coordination system.

### 2.1.1.3 Selection of the principal component subspace dimension

Once the most significant variables are incorporated into the model and the data are suitably pre-treated, the latent space dimensionality in which the data are projected have to be chosen. Suggestions about how to determine the ideal dimensionality of the reduced latent variable subspace can be found in Valle *et al.* (1999) and Li *et al.* (2002), where several methods are compared. However, the most effective methods for the choice of the latent space dimensionality are the SCREE test (Jackson, 1991) for its simplicity, the cross-validation (Wold, 1978; Eastment and Krzanowski, 1982) although it is computational demanding, and the variance reconstruction error (Qin and Dunia, 2000), which dispels some ambiguities of the previews ones. The first and the second methods are considered in this work for their simplicity.

In particular, the basic idea of cross-validation (Mosteller and Wallace, 1963) is:

- randomly dividing the data in a series of subgroups of one or more observations, cutting the  $\mathbf{X}$  matrix into a series of horizontal segments;
- building a reduced datasets deleting one of the subgroups, and calibrating the multivariate statistical model on the reduced dataset and to evaluate the goodness of fitting for the deleted subgroup through a proper criterion;
- iterating the procedure for all the subgroups;
- repeating all the previews points changing the number of retained PCs.

The criterion to estimate the goodness of fitting is to observe the root-mean-square error of cross-validation ( $RMSECV$ ), which depends on the prediction error sum of squares ( $PRESS$ ):

$$RMSECV = \sqrt{\frac{PRESS}{IJ}} \quad (2.21)$$

$$PRESS = \sum_{j=1}^J \sum_{i=1}^I (x_{i,j} - \hat{x}_{i,j})^2 = \sum_{j=1}^J \sum_{i=1}^I e_{i,j}^2, \quad (2.22)$$

with  $e_{i,j}$  element of row  $i$  and column  $j$  of the residual matrix  $\mathbf{E}$ . Adding relevant PCs should decrease the error, while adding PCs that explain only noise should increase the error. In correspondence to the minimum of  $RMSECV$ , the ideal number of PC is identified.

In the industrial practice, it is often experienced that the function of the  $RMSECV$  is monotonically decreasing with the addition of PCs. In this situation, the SCREE test is suggested (Jackson, 1991; Ku *et al.*, 1995). This is a simple, empirical, and graphical methodology that analyzes the function of the explained variance (i.e. the characteristic roots of the covariance matrix) of PCA models while changing the number of PCs. The method looks for a “knee” point in the plot of the residual percent variance against the number of PCs. The method is based on the idea that the residual variance should reach a steady state when the PCs begin to account for random errors. When a break point is found and the variance stabilizes, that point corresponds to the number of PCs which properly represent the original data.

### 2.1.2 Projection on latent structures (PLS; partial least squares regression)

PLS is a multivariate statistical method to relate two sets of data by a multivariate linear model. It was developed as a regression tool to consider the correlation between the variables of two sets of data  $\mathbf{X}$  and  $\mathbf{Y}$ . PLS exploits the typical ability of the multivariate methods to analyze many noisy and collinear data, dealing with the ill-conditioned regression problems and performing a multivariate linear regression.

Let us first consider the problem of multiple linear regression as a premise. To relate  $I$  samples of  $J$  variables collected in a matrix  $(I \times J)$  of primary variables  $\mathbf{X}$  to a matrix  $(I \times Q)$  of  $M$  secondary variable  $\mathbf{Y}$ , a linear polynomial function may be assumed:

$$\mathbf{Y} = \mathbf{XB} + \mathbf{E}, \quad (2.23)$$

with  $\mathbf{E}$  the residual vector and  $\mathbf{B}$  the matrix of regression coefficients. Three different situations arise:

- if  $J > I$  (the number of variables is higher then the one of the samples), there is a finite number of solutions;
- if  $J = I$  (the number of variable is equal to the one of the samples), a single solution is present;
- if  $J < I$  (the number of samples is higher then the one of the variables), it is impossible to find an analytical solution, but the least squares method can be used instead:

$$\mathbf{B} = (\mathbf{X}^T \mathbf{X})^{-1} \mathbf{X}^T \mathbf{Y} \quad . \quad (2.24)$$

However, because of the collinearity of the data in  $\mathbf{X}$ , the inversion of  $(\mathbf{X}^T \mathbf{X})$  is difficult, if not impossible.

To overcome the problem of collinearity, to investigate the correlation between primary variables, and to find the correlation between primary and secondary variables, PLS is a valuable help (Geladi and Kowalski, 1986; Dayal and MacGregor, 1997a). In summary, it is a method which explains the directions of maximum variability of  $\mathbf{X}$  that better predict  $\mathbf{Y}$ . In fact, PLS reduces the dimension of the system, simultaneously finding the space of LVs that are more predictive for the secondary variables and are near to the direction of maximum variability of the primary variables.

The method consists of two outer relations and one inner relation. The outer relations are:

$$\left\{ \begin{array}{l} \mathbf{X} = \mathbf{TP}^T + \mathbf{E} = \sum_{r=1}^A \mathbf{t}_r \mathbf{p}_r^T + \mathbf{E} \\ \mathbf{Y} = \mathbf{UQ}^T + \mathbf{F} = \sum_{r=1}^A \mathbf{u}_r \mathbf{q}_r^T + \mathbf{F} \end{array} \right. , \quad (2.25)$$

where  $\mathbf{t}_r$  and  $\mathbf{u}_r$  are score vectors (in  $\mathbf{T}$  and  $\mathbf{U}$ , score matrices),  $\mathbf{p}_r$  and  $\mathbf{q}_r$  are loading vectors (in  $\mathbf{P}$  and  $\mathbf{Q}$  loading matrices), and  $\mathbf{E}$  and  $\mathbf{F}$  are residual matrices whose norms  $\|\mathbf{E}\|$  e  $\|\mathbf{F}\|$  should be minimized in the least squares sense. The inner relation is a linear relation between the scores of the two matrices:

$$\mathbf{u}_r = b_r \mathbf{t}_r \quad \text{for } r = 1, \dots, R \quad , \quad (2.26)$$

where  $b_r$  are regression coefficients:

$$b_r = \frac{\mathbf{u}_r^T \mathbf{t}_r}{\mathbf{t}_r^T \mathbf{t}_r} \quad . \quad (2.27)$$

However, this is not the best algorithm from the computational point of view, because the principal components are calculated for the two blocks separately, and so exhibit a weak relation to each other. It would be preferable to give to the scores  $\mathbf{t}_r$  and  $\mathbf{u}_r$  information about each other, so that the LVs can be slightly rotated to lie closer to the secondary variables. A modified version of the NIPALS algorithm is adopted for the model building, and is described in the following subsection.

### 2.1.2.1 Non-iterative partial least squares algorithm

A modified version of the NIPALS algorithm is described here (Geladi and Kowalski, 1986), in which the scores of  $\mathbf{X}$  and  $\mathbf{Y}$  exchange information about each other, adopting some weights  $\mathbf{w}_r$  as a mathematical artifice to maintain orthogonal scores.

The algorithm develops through the following stages:

1. fix a column  $\mathbf{x}_j$  of  $\mathbf{X}$  as initial value of the score  $\mathbf{t}_1$  and fix as initial estimation of the score  $\mathbf{u}_1$  a column  $\mathbf{y}_q$  of  $\mathbf{Y}$ :

$$\begin{cases} \mathbf{t}_1 = \mathbf{x}_j \\ \mathbf{u}_1 = \mathbf{y}_q \end{cases} ; \quad (2.28)$$

2. calculate at iteration  $r$ :

$$\mathbf{w}_r = \frac{\mathbf{X}^T \mathbf{u}_r}{\|\mathbf{X}^T \mathbf{u}_r\|} ; \quad (2.29)$$

$$\mathbf{t}_r = \mathbf{X} \mathbf{w}_r ; \quad (2.30)$$

$$\mathbf{q}_r = \frac{\mathbf{u}_r^T \mathbf{t}_r}{\|\mathbf{u}_r^T \mathbf{t}_r\|} ; \quad (2.31)$$

$$\mathbf{u}_r = \mathbf{Y} \mathbf{q}_r ; \quad (2.32)$$

3. check the convergence of the values calculated through (2.30) and (2.32) to the ones of (2.28) for less than a predetermined tolerance;
4. calculate the loadings;

$$\mathbf{p}_r = \frac{\mathbf{X}^T \mathbf{t}_r}{\|\mathbf{t}_r^T \mathbf{t}_r\|} ; \quad (2.33)$$

5. update the loadings, scores, and weights:

$$\mathbf{p}_{r,\text{new}}^T = \frac{\mathbf{p}_{r,\text{new}}^T}{\|\mathbf{p}_{r,\text{old}}^T\|} ; \quad (2.34)$$

$$\mathbf{t}_{r,\text{new}} = \mathbf{t}_{r,\text{old}} \|\mathbf{p}_{r,\text{old}}\| ; \quad (2.35)$$

$$\mathbf{w}_{r,\text{new}} = \mathbf{w}_{r,\text{old}} \left\| \mathbf{p}_{r,\text{old}} \right\|^{-1} ; \quad (2.36)$$

6. compute regression coefficients by (2.27);

7. compute residuals:

$$\mathbf{E}_r = \mathbf{X}_r - \mathbf{t}_r \mathbf{p}_r^T ; \quad (2.37)$$

$$\mathbf{F}_r = \mathbf{Y}_r - b_r \mathbf{u}_r \mathbf{q}_r^T ; \quad (2.38)$$

8. repeat the procedure for every retained LV, returning to point 1 after replacing  $\mathbf{X}$  and  $\mathbf{Y}$  with  $\mathbf{E}_r$  and  $\mathbf{F}_r$ , respectively.

Scores and loadings maintain the abovementioned properties of orthogonality and orthonormality, respectively. Besides, the weights are orthonormal:

$$\mathbf{w}_r^T \mathbf{w}_s = \delta_{r,s} \left\| \mathbf{w}_r \right\|^{-1} , \quad (2.39)$$

where  $\delta_{r,s}$  is the Kronecker delta.

### 2.1.2.2 Variable selection in PLS models

For a successful calibration of multivariate statistical models the variable selection is of decisive importance. The lack of an appropriate variable selection can spoil the PLS model building, as shown by Höskuldsson (2001).

Some of the methods proposed for the variable selection are the stepwise collinearity diagnostic (Brauner and Shacham, 2000), the orthogonal scatter correction (Höskuldsson, 2001), the projection pursuit based method (Zhai *et al.*, 2006) or the bootstrapping-based variable selection (Chu *et al.*, 2004). Chong and Jun (2005) compare some of the most important methodologies, among which the most interesting are the stepwise regression and the one based on the variable importance in the projection (i.e. *VIP* index). The stepwise regression is a standard procedure which evaluates the most important variables through the effect of the sequential insertion of variables on the model. The *VIP* method, suggested by Chong and Jun (2005), demonstrates to be the most effective. The *VIP* method selects the most important variables to the sake of the PLS regression through the so-called *VIP* index:

$$VIP_j = \sqrt{J \frac{\sum_{r=1}^A b_r^2 \mathbf{t}_r^T \mathbf{t}_r \left( \frac{\mathbf{w}_r}{\|\mathbf{w}_r\|} \right)^2}{\sum_{r=1}^A b_r^2 \mathbf{t}_r^T \mathbf{t}_r}} . \quad (2.40)$$

The *VIP* index gives a measure of the variable importance in a specific PLS model evaluating the effect of the weights normalized with respect to regression coefficients and scores. In particular, the variables are deemed to be important following a rule of thumb that fixes a cut-off value at  $VIP_j=1$ . It is to say that all the variables whose  $VIP_j$  is greater than one are considered valuable predictors in the regression problem.

### 2.1.3 Monitoring charts

Once the model is calibrated, the overall conformance of a new individual observation  $\mathbf{x}_{I+1}$  to the selected optimal reference can be checked through the investigation of two parameters:

- one parameter that assesses the deviation of the new observation from the average conditions of the reference;
- one parameter that evaluates the model representativeness, namely how well the model fits the actual conditions of the new incoming observation.

These parameters are statistical indices, which are a synthetic response on the status of the new observation, and are used to build statistical monitoring charts.

The first step to evaluate the new incoming observation is to project it in the reduced space of the PCs in the case of PCA:

$$\hat{\mathbf{t}}_{I+1} = \mathbf{x}_{I+1} \mathbf{P} \quad , \quad (2.41)$$

or in the reduced space of the LVs in the case of PLS:

$$\hat{\mathbf{t}}_{I+1} = \frac{\mathbf{x}_{I+1} \mathbf{W}}{\mathbf{P}^T \mathbf{W}} \quad , \quad (2.42)$$

where the predicted score can be used for the estimation of the secondary variable:

$$\hat{\mathbf{y}}_{I+1} = \hat{\mathbf{t}}_{I+1} \mathbf{Q}^T \quad . \quad (2.43)$$

The score  $\hat{\mathbf{t}}_{I+1}$  is the projection of  $\mathbf{x}_{I+1}$  into the coordinate space of the LVs.

Furthermore, the values of all the scores  $\hat{\mathbf{t}}_{I+1}$  can be cumulated in a single number, the Hotelling  $T_{I+1}^2$  statistics:

$$T_{I+1}^2 = (\bar{\mathbf{x}}_{I+1} - \mathbf{E}(\mathbf{x}_{I+1})) \mathbf{S}^{-1} (\bar{\mathbf{x}}_{I+1} - \mathbf{E}(\mathbf{x}_{I+1}))^T = \hat{\mathbf{t}}_{I+1} \mathbf{\Lambda}^{-1} \hat{\mathbf{t}}_{I+1}^T \quad , \quad (2.44)$$

where  $\mathbf{\Lambda}$  is the diagonal matrix of the eigenvalues of  $\mathbf{S}$ , the estimation of the covariance matrix  $\mathbf{\Sigma}$  from the available data, and  $\mathbf{E}(\mathbf{x}_{I+1})$  is the expected value of  $\mathbf{x}_{I+1}$ .

In summary, the status of the new individual observation  $\mathbf{x}_{I+1}$  with respect to the average conditions imposed by the reference dataset can be assessed through the scores or through the Hotelling statistics.

However, the individual observation vector is not completely characterized in the space of the latent variables. In fact, in the score space of PCA the new observation  $\mathbf{x}_{I+1}$  is represented by:

$$\hat{\mathbf{x}}_{I+1} = \frac{\hat{\mathbf{t}}_{I+1} \mathbf{P}^T}{\mathbf{P}^T \mathbf{P}} = \hat{\mathbf{t}}_{I+1} \mathbf{P}^T \quad , \quad (2.45)$$

with an error  $\mathbf{e}_{I+1}$  of reconstruction that is given by:

$$\mathbf{e}_{I+1} = \mathbf{x}_{I+1} - \hat{\mathbf{x}}_{I+1} \quad . \quad (2.46)$$

Consequently, to the purpose of a complete characterization of the new incoming sample, the non-systematic part of the data (i.e. noise) falling into the residuals should be monitored through the residuals sum of squares of the new observation:

$$SPE_{I+1} = (\mathbf{x}_{I+1} - \hat{\mathbf{x}}_{I+1})(\mathbf{x}_{I+1} - \hat{\mathbf{x}}_{I+1})^T = \mathbf{e}_{I+1} \mathbf{e}_{I+1}^T \quad , \quad (2.47)$$

which indicates if the new observation  $\mathbf{x}_{I+1}$  can be adequately represented by the original subset of latent variables.

Since the attributes of a process must stay close to a predetermined target value without changing perceptibly, the multivariate SPC assumes that the process adheres to a state of statistical control (conformance) if it is within certain limits. Through PCA and PLS it is possible to find out statistical confidence limits that work as efficient detectors of in-control (in-spec) or out-of-control (out-of-spec) states. Confidence limits are interval estimators of population parameters, and describe the interval where it is likely to include the product/process parameters which can be considered acceptable. The confidence limits are constructed on the basis of a given dataset. If  $\theta$  is an unknown parameter from a population and  $\Theta$  the set of all the possible  $\theta$ , a confidence region  $R(\mathbf{X})$  of likely  $\theta$  values can be determined by a set of data measurements  $\mathbf{X}$ . In particular,  $R(\mathbf{X})$  is said to be a  $100(1-\alpha) \%$  confidence region when:

$$P[\theta \in R(\mathbf{X}) \Rightarrow \theta \in \Theta] = (1-\alpha) \quad , \quad (2.48)$$

namely the probability  $P$  that the parameter belongs to the population is  $(1-\alpha)$  if it is found to belong to the confidence region.

Monitoring the average value of a sample means determining whether a specific value of the average belongs plausibly to the population. In terms of hypothesis testing, monitoring the observation average is testing:

$$\begin{cases} H_0 : \bar{\mathbf{X}} = \boldsymbol{\mu}_0 \\ H_1 : \bar{\mathbf{X}} \neq \boldsymbol{\mu}_0 \end{cases} \quad (2.49)$$

where  $H_0$  is the null hypothesis,  $H_1$  is the alternative hypothesis,  $\boldsymbol{\mu}_0$  is the vector of the expected values of the  $J$  variables of the matrix  $\mathbf{X}$ , and  $\bar{\mathbf{X}}$  is the array of the mean values of the variables of  $\mathbf{X}$ .

To evaluate the confidence region for the average conditions of a  $J$ -dimensional population, the limits are calculated by:

$$P \left[ I(\bar{\mathbf{X}} - \boldsymbol{\mu}) \mathbf{S}^{-1} (\bar{\mathbf{X}} - \boldsymbol{\mu})^T \leq \frac{(I-1)J}{I-J} F_{J,I-J,\alpha} \right] , \quad (2.50)$$

where  $\mathbf{S}$  is the estimator of the covariance matrix, and the term  $(\bar{\mathbf{X}} - \boldsymbol{\mu}) \mathbf{S}^{-1} (\bar{\mathbf{X}} - \boldsymbol{\mu})^T$  has a  $T_{J,I}^2$  distribution with  $I$  and  $J$  degrees of freedom, and is related to a Fisher's F-distribution  $F_{J,I-J,\alpha}$  that is the upper  $100\alpha^{\text{th}}$  percentile of the F-distribution (Johnson and Wichern, 2007). Given this premise, the confidence limit of the Hotelling statistics at the  $100(1-\alpha)$  % level of confidence for a system of  $A$  latent variables and  $I$  samples is:

$$T_{\text{lim}}^2(A, I, \alpha) = \frac{(I-1)A}{I-A} F_{J,I-J,\alpha} , \quad (2.51)$$

which determines in the  $A$ -dimensional score space an ellipsoidal confidence region, whose semi-axis are:

$$s_r = \sqrt{\lambda_r T_{\text{lim}}^2(A, I, \alpha)} \quad \text{with } r = 1, \dots, A \quad . \quad (2.52)$$

This is a generalization on a multivariate system of the Student's t-distribution imposed to the single scores (Härdle and Simar, 2007). Indeed, for the  $r^{\text{th}}$  score, the univariate limit at  $100(1-\alpha)\%$  confidence level is:

$$t_{\text{lim}}(r, \alpha) = \pm t_{I-1, \frac{\alpha}{2}} \sqrt{\lambda_r} \quad \text{with } r = 1, \dots, A \quad . \quad (2.53)$$

where  $t_{I-1, \alpha/2}$  is the Student's t-distribution with  $(I-1)$  and  $\alpha/2$  degrees of freedom.

The components which remain unexplained in the projection to the reduced space of LVs have to be monitored, as well. The sum of squares of the unexplained components is the  $SPE_i$ , that derives from (2.47):

$$SPE_i = \sum_{j=1}^J e_{i,j}^2 \quad \text{with } i = 1, \dots, I \quad . \quad (2.54)$$

The distribution of the representation errors  $SPE_i$  can be well approximated as the distribution of  $c$  times a  $\chi^2$ -distribution (Box, 1954):

$$\left\{ \begin{array}{l} \overline{SPE} = \frac{1}{I} \sum_{i=1}^I SPE_i = cv \\ s_{SPE}^2 = \frac{1}{I-1} \sum_{i=1}^I (SPE_i - \overline{SPE})^2 = 2c^2v \end{array} \right. \Rightarrow \left\{ \begin{array}{l} c = \frac{s_{SPE}^2}{2\overline{SPE}} \\ v = 2 \frac{\overline{SPE}}{s_{SPE}^2} \end{array} \right. , \quad (2.55)$$

so the upper limit at a confidence level  $100(1-\alpha)\%$  for the  $SPE_i$  is:

$$SPE_{\lim}(\alpha) = c\chi_{v,\alpha}^2 , \quad (2.56)$$

and  $\chi_{v,\alpha}^2$  is the  $\chi^2$ -distribution with  $v$  and  $\alpha$  degrees of freedom. In this Thesis,  $SPE_{\lim}(\alpha)$  is calculated through the approximate distribution created by Jackson and Mudholkar (1979):

$$SPE_{\lim}(\alpha) = \theta_1 \left( \frac{z_\alpha \sqrt{2\theta_2 h_0^2}}{\theta_1} + 1 + \frac{\theta_2 h_0 (h_0 - 1)}{\theta_1^2} \right)^{\frac{1}{h_0}} , \quad (2.57)$$

where:

$$\theta_n = \sum_{j=A+1}^R \lambda_j^n \quad \text{for } n=1,2,3 \quad , \quad (2.58)$$

and:

$$h_0 = 1 - \frac{2\theta_1\theta_3}{3\theta_2^2} , \quad (2.59)$$

where  $z_\alpha$  is the normal standard deviate corresponding to the upper  $(1-\alpha)$  percentile.

In summary, when a new observation  $\mathbf{x}_{I+1}$  is available it can be judged by comparing it to the ones of the reference. To do this, the monitoring procedure goes through the following steps:

- projection of  $\mathbf{x}_{I+1}$  onto the sub-space of PCs or LVs as  $\hat{\mathbf{t}}_{I+1}$ ;
- comparison between the  $A$  components of the  $\hat{\mathbf{t}}_{I+1}$  and the respective confidence limits  $t_{\lim}(a, \alpha)$ , or in a more effective fashion through multivariate indices  $T_{I+1}^2$  or  $SPE_{I+1}$  observing the conditions:

$$\left\{ \begin{array}{l} T_{I+1}^2 \leq T_{\lim}^2(A, I, \alpha) \\ SPE_{I+1} \leq SPE_{\lim}(\alpha) \end{array} \right. . \quad (2.60)$$

- if the conditions (2.61) are satisfied, this means that the new observation is in a state of statistical control (conformance to the reference), with a probability of  $100(1-\alpha)\%$ , otherwise there is either a problem determined by the change in the mean conditions of the

product/process ( $T_{I+1}^2 > T_{\text{lim}}^2(A, I, \alpha)$ ) or a problem in the representativeness of the statistical model ( $SPE_{I+1} > SPE_{\text{lim}}(\alpha)$ ).

This monitoring procedure is equivalent to test the hypothesis of conformance of  $\mathbf{x}_{I+1}$  to the reference set for both  $T_{I+1}^2$  and  $SPE_{I+1}$ .

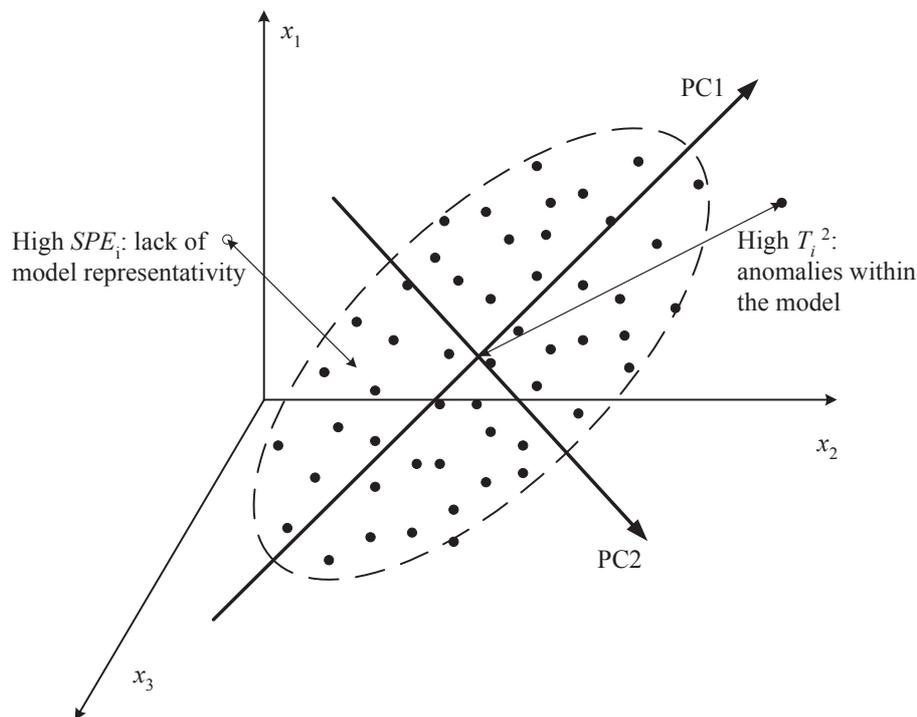
However, a word of caution regarding the use of confidence limits on scores and Hotelling statistics is advised by Wise and Gallagher (1996). In fact, the confidence limits can be found only under specified conditions. Indeed, when the hypothesis of normal and uncorrelated input data (IID, independent and identically distributed variables) is assumed, the central limit theorem<sup>1</sup> can be invoked. The assumption that a sample is drawn from a IID population is necessary to obtain the distribution of the test statistics, to build the confidence limits, and to estimate the proportion of a population that falls within certain limits (Jackson, 1991). Actually, it is possible to invoke the central limit theorem, which states that the *scores*, which are linear combinations of the original variables, derived from a sufficiently large  $\mathbf{X}$  dataset are normally distributed, only if the  $J$  variables of  $\mathbf{X}$  are IID random variables. On the contrary, if the original variables are not IID, the abovementioned fundamental assumption is violated, and the scores are not normally distributed. This determines that the confidence limits can not be valid in the aforesaid form.

Therefore, PCA and PLS models can be adequate representation of a phenomenon only if there is no data autocorrelation, the cross-correlation among variables is constant through the available samples, and the original variables are normally distributed (Kourti, 2003). In other words, the multivariate statistical techniques are successful only when *common cause* variability affects a process and when the process variables are normally distributed and independent over time or space. These conditions are rarely satisfied, and the processes products often show clear non-linear behaviour and changes in the correlation structure between variables, and in space and time.

Finally, a geometrical interpretation of PCA and PLS is shown in Figure 2.2. The samples of an optimal reference are projected from the original space onto a space of reduced dimensions made of latent variables, which are the directions of maximum variability of the data. Within this sub-space, the new observations can be analyzed through the  $T^2$  and  $SPE$  indices. In particular, the  $T^2$  index indicates the distance of the new observation from the average conditions of the reference, and the  $SPE$  indicates the distance of the new observation from the hyper-plane of latent variables.

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<sup>1</sup> The central limit theorem states that the sum of a sufficiently large number of IID random variables will tend to be normally distributed.



**Figure 2.2** Geometrical interpretation of the confidence limits in multivariate SPC and SQM.

A large value of the Hotelling statistics (i.e. the new observation is out of the elliptical limits) is an indicator of an unusual variation *within* the model, while a large  $SPE$  value (i.e. the new observation overcome the limit perpendicular distance from the hyper-plane of the latent variables) identifies anomalies *outside* the model.

### 2.1.3.1 Contribution plots, limits on the contribution plots, and relative contributions

When a new observation  $\mathbf{x}_{t+1}$  does not meet the NOC and an abnormal variation is detected by the monitoring charts, further analyses are needed to find which variable (or set of variables) causes the current state of the process (product) to be out-of-control (out-of-spec). The contribution of the  $J$  variables to the observed value of  $T_{t+1}^2$  or  $SPE_{t+1}$  helps to make a sound guess for the assignable causes of the abnormality (Nomikos, 1996). The use of contribution plots is the most common approach to detect the root cause of the problem. The contribution plots evaluate the contribution of each primary variable  $j$  to the relevant monitoring statistics, either  $T^2$  or  $SPE$ . When an anomaly is detected by the Hotelling statistics or the residuals, it is helpful to compare the contribution of every original variable to the relevant statistics with the usual value of the contribution in the NOC identified by the reference dataset. For this reason, the use of confidence bounds for the contribution to the Hotelling statistics and to the residuals were proposed (Conlin *et al.*, 2000).

The contribution  $c_{i,j}^{T^2}$  of every variable  $j$  to the  $T_i^2$  for an observation  $\mathbf{x}_i$  is determined by the square root of the Hotelling statistics of equation (2.44):

$$c_{i,j}^{T^2} = \mathbf{t}_i \mathbf{\Lambda}^{-\frac{1}{2}} \mathbf{p}_j^T \quad . \quad (2.61)$$

This is derived by the contribution  $c_{i,j}^t$  of every variable  $j$  to the scores that compose the  $T_i^2$  :

$$c_{i,j}^t = \mathbf{x}_i p_{i,j} \quad , \quad (2.62)$$

where  $\mathbf{x}_i$  is the row vector of the  $\mathbf{X}$  data matrix referring to the  $i^{\text{th}}$  observation,  $\mathbf{t}_i$  is the row vector referring to the  $i^{\text{th}}$  observation of the score matrix  $\mathbf{T}$ ,  $\mathbf{p}_j$  is the row vector referring to the  $j^{\text{th}}$  variable of the loading matrix  $\mathbf{P}$ ,  $p_{i,j}$  is an element of  $\mathbf{P}$ .

Similarly, the contribution  $c_{i,j}^E$  of every variable  $j$  to the square predicting error  $SPE_i$  of the  $i^{\text{th}}$  observation is a single element  $e_{i,j}$  of the residual matrix  $\mathbf{E}$ :

$$c_{i,j}^E = e_{i,j} \quad . \quad (2.63)$$

The values of  $c_{i,j}^{T^2}$ ,  $c_{i,j}^t$ , and  $c_{i,j}^E$  describe how each variable contributes to the Hotelling statistics, to the scores and to the residuals, respectively, and can be positive or negative (Westerhuis *et al.*, 2000).

In summary, it is possible to collect the contributions of all the  $J$  variables for all the  $I$  observations in the  $(I \times J)$   $\mathbf{X}$  matrices of the contributions to  $T^2$  and  $SPE$ :

$$\mathbf{C}^{T^2} = \left\{ c_{i,j}^{T^2} \right\} \quad i = 1, \dots, I \text{ and } j = 1, \dots, J \quad , \quad (2.64)$$

and:

$$\mathbf{C}^E = \left\{ c_{i,j}^E \right\} \quad i = 1, \dots, I \text{ and } j = 1, \dots, J \quad , \quad (2.65)$$

respectively.

Based on the assumption that both the contributions  $c_{i,j}^{T^2}$  to the  $T^2$  statistics and the contributions  $c_{i,j}^E$  to the residuals are IID, the 100(1- $\alpha$ )% confidence intervals for the contributions can be found by:

$$c_{j,\text{lim}}^{T^2}(\alpha) = \bar{c}_j^{T^2} \pm z_{\frac{\alpha}{2}} s_{\bar{c}_j^{T^2}} \quad , \quad (2.66)$$

$$c_{j,\text{lim}}^E(\alpha) = \bar{c}_j^E \pm z_{\frac{\alpha}{2}} s_{\bar{c}_j^E} \quad , \quad (2.67)$$

which are the upper (when the sign + is retained) and lower (when the sign - is retained) confidence bounds. The confidence limits of the contribution plots of equations (2.67) and (2.68) are calculated by the average contributions that a determined variable  $j$  assumes over all the  $I$  observations of the reference  $\bar{c}_j^{T^2}$  and  $\bar{c}_j^E$ :

$$\bar{c}_j^{T^2} = \frac{1}{I} \sum_{i=1}^I c_{i,j}^{T^2} \quad , \quad (2.68)$$

$$\bar{c}_j^E = \frac{1}{I} \sum_{i=1}^I c_{i,j}^E \quad , \quad (2.69)$$

and the respective standard deviations  $s_{\bar{c}_j^{T^2}}$  and  $s_{\bar{c}_j^E}$ :

$$s_{\bar{c}_j^{T^2}} = \frac{1}{I} \sum_{i=1}^I \left( c_{i,j}^{T^2} - \bar{c}_j^{T^2} \right) \quad , \quad (2.70)$$

$$s_{\bar{c}_j^E} = \frac{1}{I} \sum_{i=1}^I \left( c_{i,j}^E - \bar{c}_j^E \right) \quad , \quad (2.71)$$

where the mean values  $\bar{c}_j^{T^2}$  and  $\bar{c}_j^E$  should be zero, because they should derive from standard normal distributions.

Therefore, when the values of  $T_{I+1}^2$  or  $SPE_{I+1}$  exceed the respective confidence limits during the monitoring of a new observation  $\mathbf{x}_{I+1}$ , instead of considering the absolute value of the contributions, the *relative* size of the contribution have to be inspected (Choi and Lee, 2005), and this can be done by comparing the contribution of a single variable to the average contribution of the same variable in the reference NOC. The cause of the  $T_{I+1}^2$  or  $SPE_{I+1}$  alarm can be diagnosed by comparing the current values of the contributions  $c_{i,j}^{T^2}$  or  $c_{i,j}^E$  to the respective limits for the entire set of the original variables  $j=1, \dots, J$ . In particular, if  $T_{I+1}^2 > T_{\lim}^2(A, I, \alpha)$  and a variable  $j^*$  (with  $j^*=1, \dots, J$ ) is found to satisfy:

$$c_{I+1,j^*}^{T^2} > c_{j^*,\lim}^{T^2}(\alpha) \quad , \quad (2.72)$$

then  $j^*$  is the variable that “feels” the effect of the fault on  $T_{I+1}^2$ . In the same way, if  $SPE_{I+1} > SPE_{\lim}(\alpha)$  and:

$$c_{I+1,j^*}^E > c_{j^*,\lim}^E(\alpha) \quad , \quad (2.73)$$

for a determined  $j^*$ , the variable  $j^*$  is suspected to be the variable which mainly affected by the root cause of the anomaly. When (2.73) or (2.74) are satisfied for more then one value of  $j^*$ , this means that the effect of a certain fault distributes on different variables. This situation

can arise when the effect of the anomaly impacts on more than one variable. Otherwise, if the anomaly distributes on all the  $J$  variables, the variable with the highest contribution-to-contribution limit ratio  $c_{I+1,j}^{T^2}/c_{j,\text{lim}}^{T^2}(\alpha)$  or  $c_{I+1,j}^E/c_{j,\text{lim}}^E(\alpha)$  is the most responsible for the perturbation of the system. Therefore, interrogating the relative contributions demonstrates to be one of the most powerful methods to get a diagnosis, whenever a fault is detected (Facco, 2005; Choi and Lee, 2005).

#### 2.1.4 Enhancement for multivariate statistical methods

In Chapter 1 it was mentioned that some of the main complications that may arise when dealing with data through multivariate statistical methods are: *i*) the varying nature of the data along time or space, and *ii*) the changeable correlation structure between data. In addition to being multivariate in nature, process data are often highly auto- and cross-correlated, and often non-linear. This situation determines that data are far from being normally distributed or independent either from other variables and from observations which are neighbour in time/space. For these reasons, the time/space varying nature and the change in the correlation structure of the data have to be taken into account. Furthermore, non-normally distributed input data make the application of the abovementioned control limits in the monitoring charts impossible.

In particular, the methods described in the previews sections can be applied only to bi-dimensional matrices  $\mathbf{X}$  ( $I \times J$ ) and  $\mathbf{Y}$  ( $I \times Q$ ) of IID data. These methodologies are good mathematical representation of the relationship between variables only when the correlation between the  $J$  variables remain the same throughout the evolution of a batch. It is often the case that data have a determined order in time or space. This adds a third dimension on the data array, and the variability in the third dimension should be considered, in addition to the correlation between variables. For example, in chemical batch processes the variables are not steady state, but show time trajectories. Another example is the one of images which can be represented by matrices of light intensities (also in different spectral channels), where neighbouring data (i.e. pixels) are correlated in space. In these examples, process/product data can be collected in 3D matrices  $\underline{\mathbf{X}}$  ( $I \times J \times K_i$ ) or  $\underline{\mathbf{Y}}$  ( $I \times Q \times H_i$ ), where  $I$  different batches (or different images) are treated as different observations, while time, space or different spectral channels represent the third dimension.  $K_i$  and  $H_i$  are the number of the samples collected along time (space) for the observation  $i$ , respectively in the matrix  $\underline{\mathbf{X}}$  and  $\underline{\mathbf{Y}}$ . Correlation is present in both the direction of the variables  $j$  (cross-correlation) and the direction of the time/space samples  $k_i$  or  $h_i$  (auto-correlation).

Further complications are added when the 3D matrices  $\underline{\mathbf{X}}$  and  $\underline{\mathbf{Y}}$  have irregular shape, due to the differences in the number of samples ( $k_i$  or  $h_i$ ) taken in time (or space). Moreover, the time trajectories of different processes variables or quality variables are sometimes not

synchronized between the  $I$  batches, or the spatial characteristics of an image are not aligned between the  $I$  observations, so  $K_r \neq K_s$  and  $H_r \neq H_s$  with  $r \neq s$ , and  $r=1, \dots, I$  and  $s=1, \dots, I$ .

To deal with the changeable nature of the correlation structure between data and the varying nature of the data, multi-way multivariate statistical techniques are commonly used.

#### 2.1.4.1 Multi-way methods, data unfolding and data synchronization/alignment

When the third dimension (i.e. time or space), is present in the data, and when the data collected in an ordered manner are assembled in regular 3D matrices  $\underline{\mathbf{X}}$  ( $I$  observations  $\times$   $J$  variables  $\times$   $K$  samples), the multi-way SPC (Nomikos and MacGregor, 1994) demonstrated to be a very effective strategy. Multi-way PCA (MPCA) and multi-way PLS (MPLS) are consistent with PCA and PLS, respectively, from both the mathematical and the algorithmic point of view. In fact, MPCA/MPLS have the same aims and benefits of PCA/PLS, because they are equivalent to perform PCA/PLS on enlarged 2D matrices derived by unfolding the 3D data matrices:

$$\underline{\mathbf{X}} = \sum_{r=1}^A \mathbf{t}_r \mathbf{P}_r + \underline{\mathbf{E}} \quad , \quad (2.74)$$

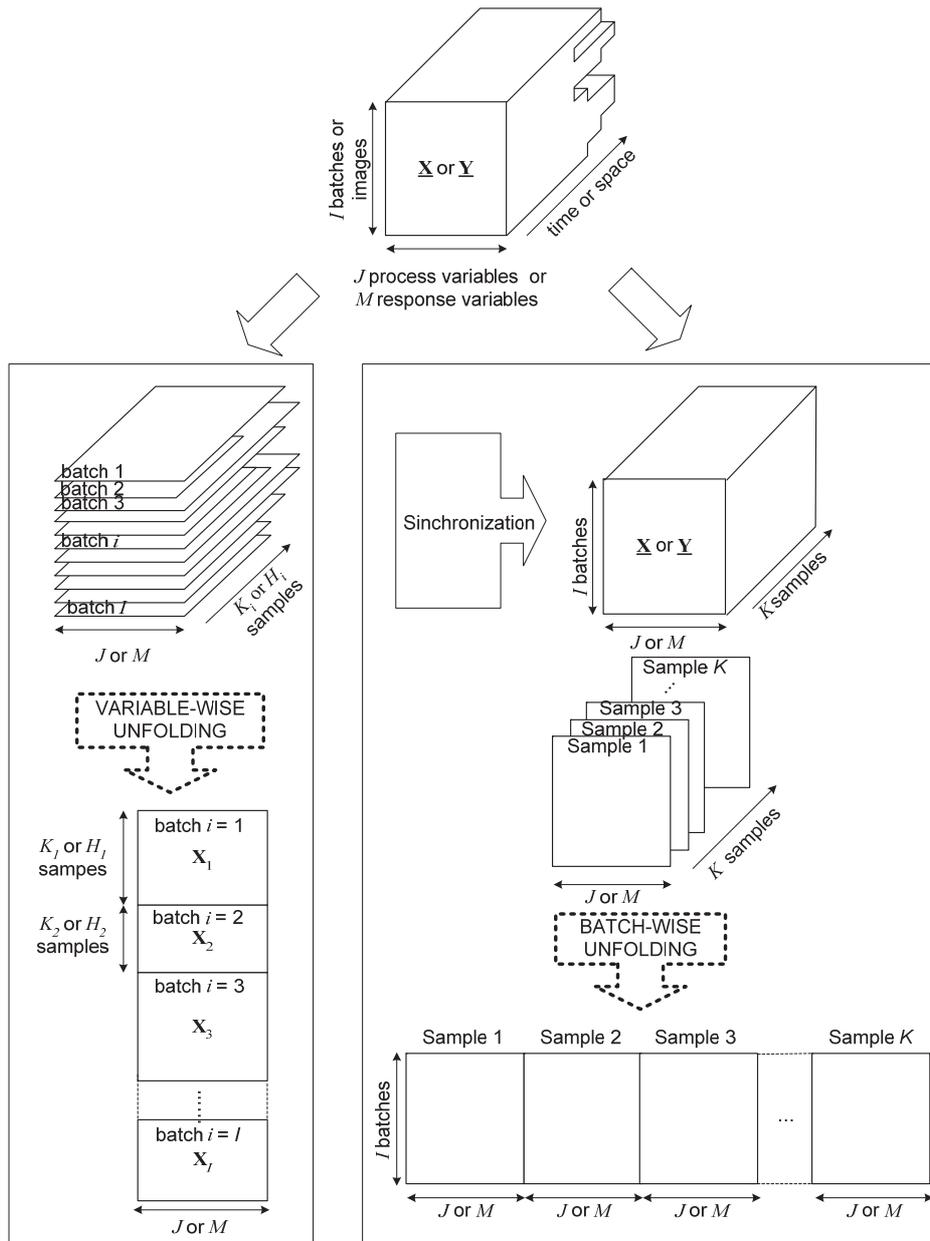
where the  $\mathbf{t}_r$ 's are the score vectors, and the  $\mathbf{P}_r$ 's are the loading matrices of the loading for all the  $J$  variables and all the  $K$  samples, the direction of maximum variability for every variable in every sample in time or space, and  $\underline{\mathbf{E}}$  is the residual matrix.

“Unfolding” is a technique to derive 2D matrices by spreading out the original 3D matrices in a meaningful way to highlight the relevant variability to be inspected. Different unfolding methods were developed (Kourti, 2003), corresponding to different ways to unfold the 3D matrix, but two of them are the most significant (Figure 2.3):

- batch-wise unfolding (BWU);
- variable-wise unfolding (VWU).

The BWU unfolding spreads out the 3D data in 2D matrices  $\mathbf{X}^{\text{BWU}}$  that consider the time/space order of the data (Wise and Gallagher, 1996), putting the time slices of the original 3D matrix side-by-side along the direction of the batches. Considering a 3D matrix  $\underline{\mathbf{X}} = \{x_{i,j,k}\}$  with  $i=1, \dots, I$ ,  $j=1, \dots, J$ ,  $k=1, \dots, K$ , where:

- the  $i^{\text{th}}$  horizontal slice  $\mathbf{X}_i$  is the matrix of the trajectories of all the  $J$  variables in all the  $K$  samples in time or space for the observation (i.e. batch or image)  $i$ ;
- the  $j^{\text{th}}$  vertical slice  $\mathbf{X}_j$  is the matrix of the time/space evolution of the variable  $j$  for all the samples  $K$  and all the observations  $I$ ;
- the  $k^{\text{th}}$  vertical slice  $\mathbf{X}_k$  is the matrix of the time/space sample  $k$  for all the  $J$  variables and all the  $I$  observations.



**Figure 2.3** Unfolding of the three-dimensional data matrix in both the variable-wise direction and the batch-wise direction.

MPCA and MPLS can be performed using PCA and PLS respectively on the batch wise unfolded 2D matrix:

$$\mathbf{X}^{\text{BWU}} = [\mathbf{X}_{k=1} \mathbf{X}_{k=2} \dots \mathbf{X}_{k=K}] \quad , \quad (2.75)$$

which is a  $(I \times JK)$  matrix.

Mean-centring the batch wise unfolded data matrix (i.e., subtracting the mean trajectory of each variable) removes the major non-linearity of the input variables (Nomikos and MacGregor, 1995b), summarizing the variability of the variables with respect to both the variables and their time/space evolution (Kourti and MacGregor, 1995). Accordingly, the

cross-correlation between variables is analyzed together with the auto-correlation in time/space within each variable. This means that, in the example of batch processes, the entire history of the batch is taken into account and the batch dynamics is properly represented into the model. In the example of the images, the spatial structure (or different spectral channels) are considered throughout the entire image in the BWU.

Anyway, some difficulties arise in the realtime application of BWU to the case of batch processes, because data are collected sequentially and are available for the entire batch only *after* the completion of the batch itself. In fact, BWU is successfully applied to run-to-run monitoring and control strategies. However, when online applications are required some issues have to be faced. In fact, before batch completion BWU works well only if at least 10% of the batch history is already available (Nomikos and MacGregor, 1995b). Furthermore, BWU presents two main limitations about the data collected in real time:

- data are often not synchronized-aligned;
- data are not available for the entire batch to perform a sequential test during a batch run.

The latter problem can be solved filling the incomplete matrix for the future unknown samples under three alternative hypothesis (Nomikos and MacGregor, 1995a):

- the future samples conform to the mean reference conditions;
- the current deviation from the mean variables' trajectory remain unchanged for the rest of the batch duration;
- using the ability of the PCA and PLS to handle missing data. The abovementioned methods to treat missing data can be used to this purpose.

The synchronization of batches of uneven duration can be a very demanding issue. Using MPCA or MPLS on batch-wise unfolded data requires effective methods for the alignment/synchronization of the variables image features or time trajectories, stretching or shrinking the actual observation to the length of a reference one. The most popular synchronization methods are:

- the dynamic time warping (Kassidas *et al.*, 1999);
- the indicator variable (Westerhuis *et al.*, 1999).

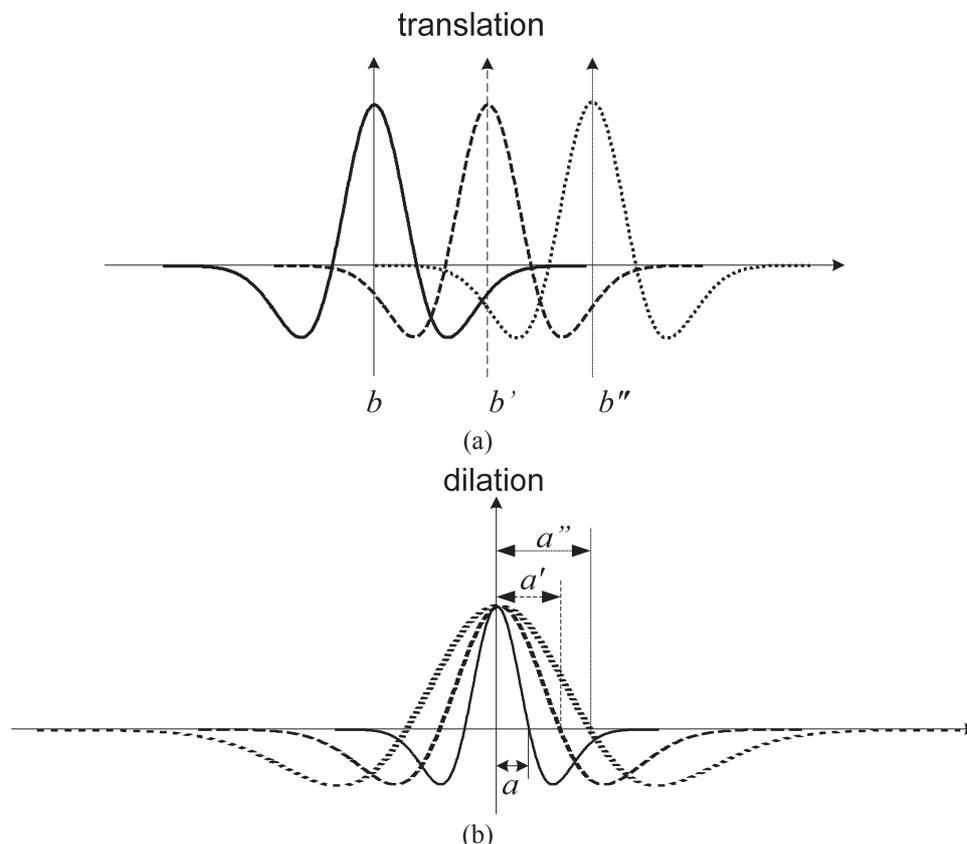
The VWU (Wold *et al.*, 1987) represents the data in 2D matrices  $\mathbf{X}^{\text{VWU}}$  ( $IK \times J$ ) that preserve the direction of the variables (Eriksson *et al.*, 2001) and do not consider the data time or space order, because they are constituted putting the slices of the observations  $\mathbf{X}_i$  of the original 3D matrix in vertical position one underneath the other:

$$\mathbf{X}^{\text{VWU}} = \begin{bmatrix} \mathbf{X}_{i=1} \\ \mathbf{X}_{i=2} \\ \vdots \\ \mathbf{X}_{i=l} \end{bmatrix} . \quad (2.76)$$

Using this procedure, it is neither necessary to estimate the future unknown part of the batch, nor to synchronize or to align the signals. The VWU approach is easier to implement online. However, if the variables do not consider the time/space order, the dynamics of the data is lost in batch processing, and the effect of the neighbourhood is lost in images. In summary, the auto-correlations are not considered in VWU. Furthermore, the VWU forces the correlation structure between data to be constant within the entire batch or image (Kourti, 2003). But considering a fixed and unchangeable correlation structure of the data is too a restrictive condition. Considering auto-correlation and the change of cross-correlation during time is the main difficulty of the VWU scheme.

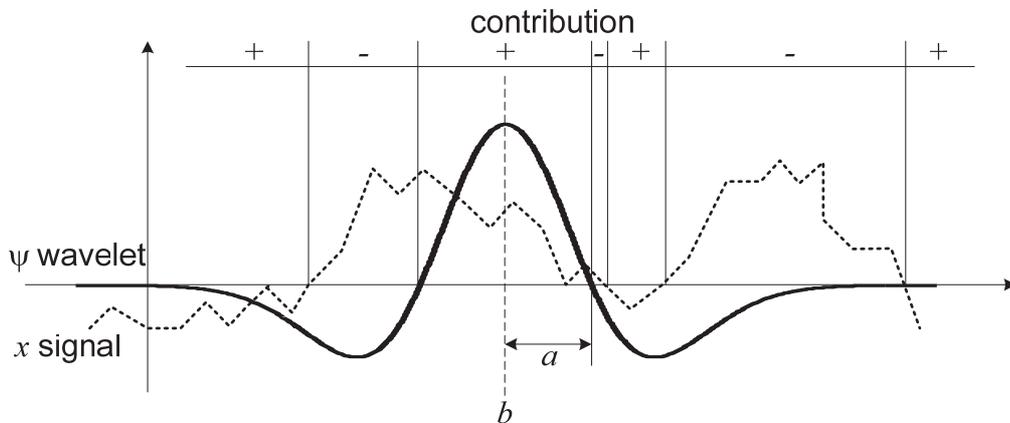
## 2.2 Multiresolution decomposition methods

Multiresolution decomposition methods are techniques which transform a signal into a representation that is more useful and easily manageable (Addison, 2002). To perform this decomposition a transformation process is needed: the wavelet transform. This procedure entails the use of wavelet functions, which are localized waveforms that spread out the signal from the original domain to the domain of frequency. This means that it is possible to convert the signal in a series of profiles, which are more linear and more normally distributed.



**Figure 2.4** Example of “Mexican hat” wavelet (a) location in the domain and (b) dilation of the scale.

The wavelet transform mechanism entails the comparison between a wavelet of scale  $a$  and location  $b$  and an arbitrary signal  $x$ . To carry out a proper decomposition, the waveforms can be translated varying its location  $b$  (moved along the domain in which it is defined, i.e. time or space) or dilated varying its size  $a$  (shrinking or widening the wavelet) (Figure 2.4). The transform results in a positive contribution when the signal and the wavelet are both positive or both negative, while the transform is negative if the signal and the wavelet are of opposite signs (Figure 2.5). The higher the correlation between signal and wavelet is, the higher the absolute value of the transform is.



**Figure 2.5** Mechanism of transformation of a signal through wavelet transform.

This means that, when the signal trajectory has approximately the same shape and size of the wavelet profile in a determined location, the transform produce a large positive value, and *vice versa* when signal and wavelet are out-of-phase. As a consequence, the smallest size of the wavelet are correlated to the highest frequencies of the signal (i.e., noise), while the widest size of the wavelet are related to the long-term fluctuations of the signal, such as drifts or seasonal effects.

Note that an advantage in the choice of multiresolution techniques is that the signatures of a signal in its domain (i.e. time or space) are maintained in the transformation of the signal from the original domain to the frequency domain. In fact, the transformed signal can be rebuilt preserving the time/space information (which is unfeasible, for example, in the Fourier transform).

In the following sections the mathematical and algorithmic aspects of the wavelet transform are presented, together with their main applications.

### 2.2.1 Continuous and discrete wavelet transform

In mathematical terms, the wavelet transform is the convolution of a signal  $x$  with  $\psi_{a,b}^*(s)$ , the complex conjugate of a “mother” wavelet function  $\psi_{a,b}$ , integrated over the signal range:

$$T(a,b) = \int_{-\infty}^{+\infty} x(s) \psi_{a,b}^*(s) ds \quad . \quad (2.77)$$

where  $s \in \mathfrak{R}$  identifies the domain. The localized and normalized waveform of the mother wavelet is:

$$\psi_{a,b}(s) = \frac{1}{\sqrt{a}} \psi\left(\frac{s-b}{a}\right) \quad , \quad (2.78)$$

where  $a$  is a dilation parameter and  $b$  a location parameter. These family of translations and dilations is a basis of the Hilbert space of square integrable functions  $L^2(\mathfrak{R})$ .

The transformation procedure compare the signal to the mother wavelet, shifting its location  $b$  and shrinking or stretching the scale  $a$ . If the signal and the wavelet are both positive or both negative in the original domain, the transform will be positive, otherwise it will be negative.

The practical implementation of the wavelet transform entails the discretization of the scales  $a$  and of the step size between different locations  $b$ . The discretization can be given by:

$$\psi_{m,n}(s) = \frac{1}{\sqrt{a_0^m}} \psi\left(\frac{s - nb_0 a_0^m}{a_0^m}\right) \quad , \quad (2.79)$$

where  $n$  and  $m$  are integer parameters which respectively control the wavelet dilation and translation. The size of the translation step is  $\Delta b = b_0 a_0^m$  and the transform becomes:

$$T_{m,n} = \int_{-\infty}^{+\infty} x(s) \frac{1}{a_0^{m/2}} \psi(a_0^{-m} s - nb_0) ds = \langle x, \psi_{m,n} \rangle \quad . \quad (2.80)$$

The inner products of  $x$  and  $\psi_{m,n}$  are called detail coefficients  $T_{m,n}$ . The simplest and most efficient discretization is the so called dyadic grid. It generates orthonormal wavelets, where  $a_0=2$  and  $b_0=1$ :

$$\psi_{m,n}(s) = 2^{-m/2} \psi(2^{-m} s - n) \quad . \quad (2.81)$$

In a discretized wavelet transform there is a finite number of wavelet coefficients, which require the evaluation of an integral. After having passed the signal through the abovementioned high-pass filter  $\psi_{m,n}$ , another function  $\phi_{m,n}$  (the so called ‘‘father’’ wavelet) is needed to avoid the numerical complication. The father wavelet have the same form as the mother wavelet:

$$\phi_{m,n}(s) = 2^{-m/2} \phi(2^{-m} s - n) \quad , \quad (2.82)$$

which are orthogonal to its translation, but not to its dilation, and performs a low-pass filter, i.e. a scaling function, which establishes the multiresolution features of the wavelet decomposition. The convolution of the scaling function with the signal produces the approximation coefficients:

$$S_{m,n} = \int_{-\infty}^{+\infty} x(s) \frac{1}{a_0^{m/2}} \varphi(a_0^{-m}s - nb_0) ds \quad , \quad (2.83)$$

so that the continuous approximation of the signal at the scale  $m$  can be generated by summing a sequence of scaling function at the scale factored by the approximation coefficients:

$$x_m(s) = \sum_{n=-\infty}^{+\infty} S_{m,n} \varphi_{n,m}(s) \quad . \quad (2.84)$$

This is an approximated, smoothed version of the original signal.

Also the original signal can be rebuilt following the reconstruction representation of the inverse wavelet transform:

$$x_{m-1}(s) = x_m(s) + d_m(s) \quad . \quad (2.85)$$

The reconstruction has no redundancy, because of the normality of the wavelet. The term  $d_m(s)$  is constituted of the detail coefficients at scale  $m$ :

$$d_m(s) = \sum_{n=-\infty}^{+\infty} T_{m,n} \psi_{m,n}(s) \quad . \quad (2.86)$$

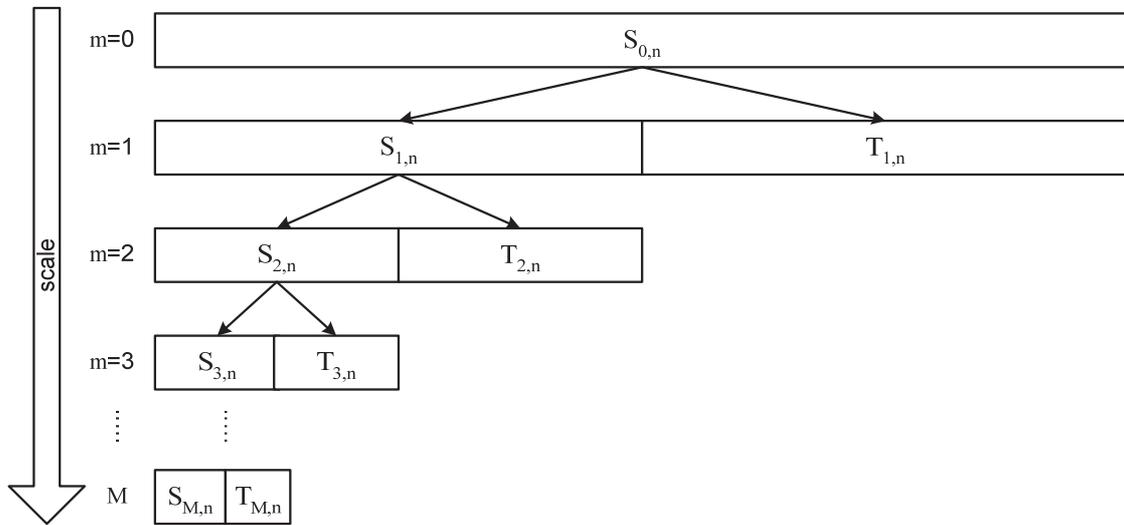
The result is that a signal can be represented combining the approximation coefficients and the series expansion of the details:

$$x(s) = \sum_{n=-\infty}^{+\infty} S_{M,n} \phi_{M,n}(s) + \sum_{m=-\infty}^M \sum_{n=-\infty}^{+\infty} T_{m,n} \psi_{m,n}(s) \quad (2.87)$$

where  $M$  is an index of the chosen scale. For a signal of finite length  $N_x$ ,  $M = \log_2 N_x$  is the maximum number of scales which can be investigated with the dyadic grid discretization.

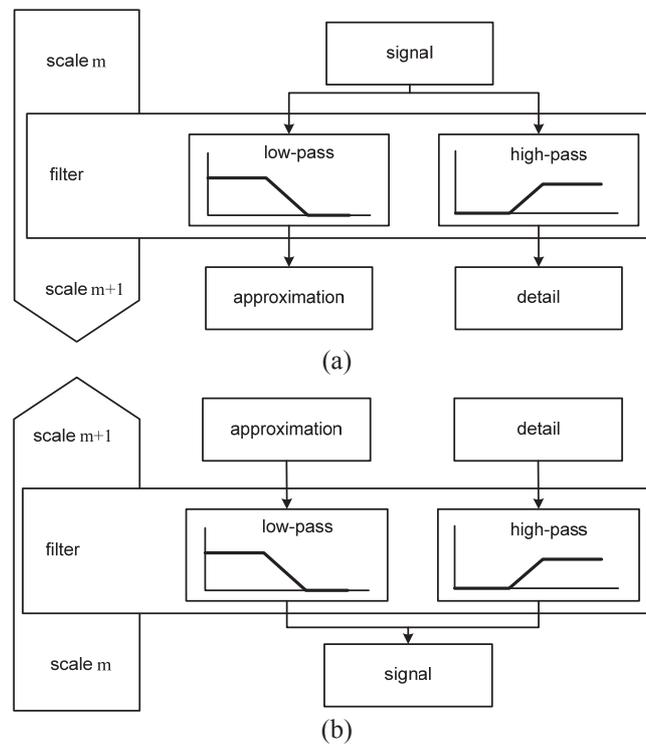
In summary, the wavelet transform is a band-pass filter, which allows the components within a predefined and finite range of frequency to relapse into the detail coefficients at each scale (Addison, 2002). Namely, at each scale the original signal is increasingly cleansed by the higher frequency components, by means of two complementary filters: a low-pass filter and a high-pass one. From the numerical point of view, the wavelet pyramidal algorithm (Mallat, 1989) decomposes a signal trajectory sequentially in a series of approximated versions of the

profile (lower frequency scales) and details (higher frequency scales), iterating the procedure at every decomposition level.



**Figure 2.6** Schematic diagram of the algorithm for the wavelet transform filtering.

Passing through the filters, the original signal is split into two parts: the approximation (which retains the high scale and low frequency part of the signal), and the detail (which summarizes the high frequency, low scale part). In this way the original signal can be studied at different resolution scales, or denoised and detrended in a meaningful manner.



**Figure 2.7** Wavelet signal filtering: (a) down-sampling associated to the signal wavelet filtering and (b) up-sampling associated to the signal reconstruction from approximations and details.

Note that, when the signal is convolved with a low-pass filter (moving the filter along the signal step-by-step of the discretized domain) a dyadic down-sampling is applied: the signal is down-sampled by a factor 2 generating the approximation, that contains the odd elements of the signal. The signal is also convolved with a high-pass filter and down-sampled to form the detail that contains the even elements of the signal (Figure 2.7).

The down-sampling retains only the odd elements of the for the approximation only the and for the detail only. This means that the approximation and the detail at scale  $m+1$  are half of the dimension of the signal at scale  $m$ . In the signal reconstruction from scale  $m$  to scale  $m+1$ , the filtering process is reversed, feeding back the larger scales components (approximations and details) through the filter, which up-samples the scales components approximation and detail and assemble them re-building the original signal. In mathematical terms this is an inverse wavelet transform.

Different types of wavelet are available for signal transformation: mexican hat wavelet (the second derivative of a Gaussian distribution function); Haar wavelet (the most effective for the representation of the discontinuities); Daubachies wavelets (the most frequently used in the texture analysis; Salari and Ling, 1997); etc.... What can be inferred by the literature is that the selection of the most proper wavelet is case sensitive, but it is suggested (Ruttiman *et al.*, 1998) the use of wavelets that:

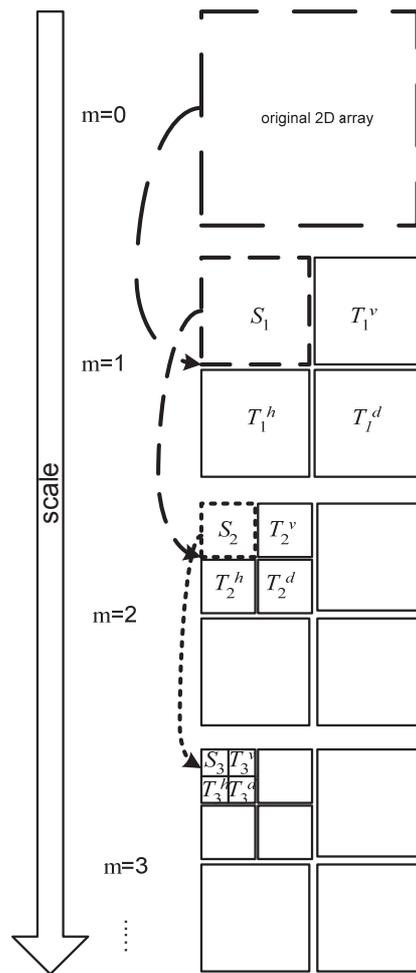
- determine limited phase distortion;
- maintain a faithful localization on the domain;
- de-correlate the signal in a sensitive manner for both the smooth features and discontinuities.

Also the choice of the proper decomposition scale is case sensitive. However, some general methodologies to select the most relevant scales are available in literature, such as the comparison of some statistical indices in different scales of resolution derived by the signals and the respective approximations (moments, entropy, skewness, kurtosis, etc...; Addison, 2002).

### 2.2.1.1 Bi-dimensional wavelet transform

In many applications (e.g. image analysis) the dataset is a 2D matrix in the domain of the variables  $\mathbf{s} \in \mathfrak{R}^2$  (Mallat, 1989). The wavelet transform can be used either to compress the data in a meaningful manner, or to perform a multiresolution characterization of the matrix. In both the cases, two-dimensional wavelet transforms are required.

The two-dimensional wavelet transforms can be generated by the tensor product of their mono-dimensional orthonormal counterparts (Addison, 2002), using the same scaling procedure as the one-dimensional scale on both the rows and the columns of the data matrix.



**Figure 2.8** Schematic diagram of the matrix manipulation to decompose the 2D array on a bi-dimensional grid through wavelet transform.

Two-dimensional scaling and wavelet functions can be defined as:

- 2D scaling function:

$$\varphi(\mathbf{s}) = \varphi(s_1)\varphi(s_2) \quad ; \quad (2.88)$$

- 2D horizontal wavelet (in the sense of the rows):

$$\psi^h(\mathbf{s}) = \varphi(s_1)\psi(s_2) \quad ; \quad (2.89)$$

- 2D vertical wavelet (in the sense of the columns):

$$\psi^v(\mathbf{s}) = \psi(s_1)\varphi(s_2) \quad ; \quad (2.90)$$

- 2D diagonal wavelet:

$$\psi^d(\mathbf{s}) = \psi(s_1)\psi(s_2) \quad ; \quad (2.91)$$

where  $s_1$  and  $s_2$  are elements of the 2D domain defined by all the  $\mathbf{s} \in \mathfrak{R}^2$  (e.g., spatial coordinates of images). Accordingly, the multiresolution decomposition can be expressed as:

$$\begin{cases} S_{m+1,(n_1,n_2)} = \frac{1}{2} \sum_{k_1} \sum_{k_2} c_{k_1} c_{k_2} S_{m,(2n_1+k_1,2n_2+k_2)} \\ T_{m+1,(n_1,n_2)}^h = \frac{1}{2} \sum_{k_1} \sum_{k_2} b_{k_1} c_{k_2} S_{m,(2n_1+k_1,2n_2+k_2)} \\ T_{m+1,(n_1,n_2)}^v = \frac{1}{2} \sum_{k_1} \sum_{k_2} c_{k_1} b_{k_2} S_{m,(2n_1+k_1,2n_2+k_2)} \\ T_{m+1,(n_1,n_2)}^d = \frac{1}{2} \sum_{k_1} \sum_{k_2} b_{k_1} b_{k_2} S_{m,(2n_1+k_1,2n_2+k_2)} \end{cases} \quad (2.92)$$

where  $k_1$  and  $k_2$  are scaling coefficients and  $n_1$  and  $n_2$  are location indices.

The general idea of a 2D wavelet decomposition is shown in Figure 2.8. After the first decomposition, the original data matrix  $\mathbf{X}_0$  is split into four distinct sub-matrices: an approximation  $\mathbf{S}_1$ ; an horizontal detail  $\mathbf{T}_1^h$ ; a vertical detail  $\mathbf{T}_1^v$ ; and a diagonal detail  $\mathbf{T}_1^d$ . In the next decomposition scale, the details are left untouched, and the next iteration decomposes only the approximation  $\mathbf{S}_1$ . The transformation at scale  $m=2$  decomposes  $\mathbf{S}_1$  in a new approximation  $\mathbf{S}_2$  and the details  $\mathbf{T}_2^h$ ,  $\mathbf{T}_2^v$  and  $\mathbf{T}_2^d$ . This procedure can be iterated  $M$  times for a  $(2^M \times 2^M)$  matrix, where the dimension of the matrices  $\mathbf{S}_m$ ,  $\mathbf{T}_m^h$ ,  $\mathbf{T}_m^v$  and  $\mathbf{T}_m^d$  is down-sampled to  $(2^{M-m} \times 2^{M-m})$ .

Once more, the original matrix can be reconstructed as:

$$\mathbf{X}_0 = \mathbf{X}_M + \sum_{m=1}^M (\mathbf{D}_m^h + \mathbf{D}_m^v + \mathbf{D}_m^d) \quad , \quad (2.93)$$

where the matrix  $\mathbf{X}_M$  is the smooth version of the original matrix at the largest scale index  $M$ , while the  $\mathbf{D}_m^h$ ,  $\mathbf{D}_m^v$  and  $\mathbf{D}_m^d$  are the reconstruction of the details from the coefficients in  $\mathbf{T}_m^h$ ,  $\mathbf{T}_m^v$  and  $\mathbf{T}_m^d$ , respectively.

