RANK DETERMINATION AND ANALYSIS OF NON-LINEAR PROCESSES BY GLOBAL LINEARIZING TRANSFORMATION

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SUMMARY

A method of rank determination which can also be used to construct models for some non-linear processes by means of a global linearizing transformation (GLT) is presented. A test case using the theoretical properties of an ideal gas is analyzed for comparison with the log linearization. A real data set from a Taguchi sensor array is analyzed by the method and the results are compared with previous work. The linearizing transformations are constructed using spline functions for the examples treated in this paper, although other kinds of functions can be used.

KEY WORDS rank determination; non-linear processes; linearizing transformations; modelling; GLT

INTRODUCTION

The use of computers to analyze chemical data has increased dramatically over the past 20 years, partially owing to the vast computational resources made available by the recent advances in hardware and software. A large scientific calculation that would be expensive and take hours on a mainframe 15 years ago can now be easily accomplished with a minicomputer, allowing some previously unthinkable problems to be solved routinely.

On the other hand, data acquisition in analytical chemistry has also reached a very sophisticated point, especially with computerized instrumentation¹ which can yield enormous quantities of very reliable chemical data. One of the most interesting features of modern instrumentation is the number of variables which can be measured on a single sample. For example, absorption intensities at 1000 or more wavelengths are commonly recorded in one sample spectrum, but of course a huge raw list of numbers is of no value for the understanding of the problem and one can easily become overwhelmed by it. Visualization is the most

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important cognitive interface between researcher and data and that also has been aided tremendously by the computer.² In many cases visualization may not be adequate without a theoretical framework to interpret the display. Consequently, the demand for more sophisticated tools to be used in the interpretation of such data has rapidly increased.

The fundamental mathematical technique supporting the majority of the existing methods for dealing with multivariate analysis is principal component analysis (PCA)^{3,4} via singular value decomposition (SVD). Beginning with data obtained from P types of measurements (P variables) on each sample, the total data collected for N samples are stored in a large $(N \times P)$ matrix **R** which carries, besides some noise, a high degree of redundancy of information due to correlated variables. PCA is motivated by the idea that the measured variables might be associated in such a way that the essential information can be reconstructed from a much smaller set of variables, $K \ll P$, which are obtained from the original data. This new compressed set of variables should contain all the important information of the original data set but arranged in an easier way to handle and interpret. In the case where K is really small, the advantage of a graphical visualization becomes much clearer.

However, it is not always perceived that in the background of the above argument, as in many other places in science,^{5,6} it is tacitly assumed that the involved variables are the result of linear functional relations. For example, in spectroscopy the theoretical model is given by Beer's law¹ in which the absorption intensities are proportional to the concentrations. Thus, theoretically, the matrix **R**, which in this case is formed by the measurements **R**_{ij} of the *j*th absorption intensity for the *i*th sample, should exhibit a rank equal to the number of different components in all samples. Deviations from the linear theory and noise from experimental errors will cause the matrix **R** to have full rank with probability one and its theoretical rank (the so-called pseudorank) must be recovered by a singular value type of analysis.

A linear functional relation seldom holds as an adequate theoretical model over a wide range of measurements. Most classical linear theoretical models in physics and chemistry are obtained from a local linearization of a more general non-linear model or result directly from linear approximate assumptions. A local linearization is always possible if the variables are smoothly related even though its restriction may be so strong as to make the approximation useless. The very same Beer's law, for instance, is an excellent approximation in a certain range of concentrations and wavelengths, but for a wider range a non-linear modification must be made. Consequently, linear methods are not always sufficient to properly analyze experimental data which are frequently collected in a wide range. In many cases the variables are highly non-linear⁷ and the linear approximation might be completely useless as a reasonable assumption for any considerable subset of the data. Thus, the development of appropriate methods is necessary for the treatment of non-linear processes.

Of course, the term 'non-linear process' is too vague to admit a unified and general treatment and any useful method for the study of processes which are not amenable to linear methods' must also have a restricted range of applications. Many of the so called 'non-linear methods' are in fact techniques which are used to reduce a (vaguely defined) class of problems into a form where usual linear methods can be employed. Locally weighted regression (LWR), for example, uses restricted regions where linear approximations are acceptable in order to cover a larger range of interest.⁸ Another way to extend the linear hypothesis is to make powers and products of independent variables take the role of new variables, which is very useful when those are in fact (at least approximately) the kind of non-linearities present in the process.⁹ The main disadvantage of this procedure is that the rank of the new data may not reflect the real dimensional structure of the process. A more general approach is to attempt a global transformation of the response variables by monotone functions so as to yield a linear process in the new transformed variables. A blend of the two previous approaches can also be employed in some cases.

For example, when analyzing a binary mixture of gases by MOS gas sensors, the *j*th probe response can be theoretically described by the function

$$R_i(c_1, c_2) = [A_i(c_1)^{m_1} + B_i(c_2)^{m_2}]^{n_j}$$
(1)

where c_1 and c_2 are the gas concentrations, A_j , B_j and nj are probe parameters and m_1 and m_2 are constants depending on the respective gases.¹⁰ If the transformations given by the functions

$$T_i(x) = x^{1/n_i} \tag{2}$$

are applied to the probe response R_j , the new response variables $Y_j = T_j(R_j)$ will be given as linear functions of the independent variables $(c_1)^{m_1}$ and $(c_2)^{m_2}$. Now, if for a set of samples $(1 \le i \le N)$ a matrix of measurements \mathbf{R}_{ij} is produced, then the transformed matrix $\mathbf{Y}_{ij} = T_j(\mathbf{R}_{ij})$ will certainly exhibit the correct pseudorank of two, since it is a collection of N vectors in \mathbb{R}^P (*i.e. the real P*-dimensional Euclidean space) obtained by a linear transformation of a collection of N vectors

$$\mathbf{c}_{i} = ((c_{2i})^{m_{1}}, (c_{2i})^{m_{2}}) \text{ from } \mathbf{R}^{2}$$
 (3)

The log function is the most commonly used linearizing transformation in a variety of applications from psychology,¹¹ chemistry,¹² biology,^{13,14} physics,¹⁵ etc., where the theoretical model relating response variables to predictor variables is given by products and powers. The Box–Cox transformation given by functions of the type

$$T(x) = \frac{e^{\lambda \log x} - 1}{\lambda}, \quad \lambda > 0$$
(4)

is also commonly used and somewhat generalizes the log transformation, which is its limit for λ approaching zero. However, it is still a very specific transformation which can be very useful in many particular instances (as in the above example), but, just like the log transformation, it cannot linearize in a general sense.

An example of useful log linearization (which will be treated as an illustration later) can be given by the study of thermodynamical properties of an ideal gas. Suppose that temperature (T) and pressure (P) are the state variables and that their values and volume (V), among other properties, are measured for a set of samples. Plotting the experimental results in a T, P versus V plot, one finds that their values for all samples lie in a 'noisy' neighborhood of a surface (Figure 1) given by the theoretical model for the ideal gas,

$$V = \frac{RT}{P}$$
(5)

If the measured temperature and pressure vary in a small range, a tangent or secant plane will be an acceptable model to describe the results. However, if the range spreads over a wider region which includes a non-negligible variation in the surface curvature, a secant plane is no longer appropriate. Now, if the log function is used to transform each variable, the theoretical model is easily expressed as a linear relation among the transformed variables; i.e.

$$P^* = \log P$$

$$T^* = \log T$$

$$V^* = \log V = \log R + \log T - \log P = C + T^* + P^*$$
(6)

 $V^* = \log V = \log R + \log T - \log P = C + T^* + P^*$ and then the values for V^* will lie in a noisy neighborhood of the plane described by T^* and P^*



Figure 1. P-V-T surface for an ideal gas

according to the above equation, which will be apparent by a PCA analysis of the data.

However, one is not always so fortunate as to be confronted with such simple theoretical relations. The log function would not be as helpful if the functional relationship between the variables included sums or other forms inappropriate to the use of logarithm properties. Suppose, for example, that the gas samples being analyzed are not under ideal conditions but behave as van der Waals gases. Then the theoretical model which relates V (and other properties) to T and P becomes much more complicated as shown by the implicit relation

$$P = \frac{RT}{V-b} - \frac{a}{V^2} \tag{7}$$

and the log transformation (or any of the above-mentioned transformations) cannot be justified.

Of course, this only means that those are not the appropriate transformations for this case, but still the main idea behind the method can be further generalized by the use of a larger class of functions. How to choose the appropriate transformation is the main question. One possible approach is to analyze a theoretical model (if there is a reliable one) to help find functions that could be good candidates to disentangle the non-linear functional relations. This depends heavily on a precise theoretical formulation of the process. A second path, followed by the present work, is to look for a global linearizing transformation (GLT) in a set of functions described by a finite number of parameters, which, although not quite 'functionally correct', is convenient owing to a balance between descriptive power and parsimony.

Many classes of functions (orthogonal polynomials, quasi-polynomials, trigonometric functions, Padé approximations, wavelets, Bézier curves, etc.) could possibly be used depending on the known special features of the problem.⁶Polynomials of some order would be an obvious first choice. On second thought, however, one quickly realizes that they are somewhat unbounded as functional construction blocks. A much more powerful and manageable class of functions can be constructed by piecewise polynomials of low order as the spline functions. In the present paper, splines^{9,17,18} are employed since they are known for their effectiveness as approximates and also because of their simple construction from a fixed basis. The splines are not used in this instance for fitting nor with the objective of approximating a

definite function, but to construct one out of many possible approximate linearizations. It must be emphasized that linearizing transformation functions are not unique and so the simpler the function used, the better.

LINEARIZING TRANSFORMATION

In a linear theoretical model in chemometrics, when samples i $(1 \le i \le N)$ are analyzed by sensors j $(1 \le j \le P)$, it is assumed that the results \mathbf{R}_{ij} of the measurements are linear functions of the concentrations of the substances present in the samples. If the sensors measure distinct properties, then each row vector of the data matrix $(\mathbf{R}_{ij}) = \mathbf{R}$ is the result of a linear function of the concentration vector $\mathbf{c}_i \in \mathbb{R}^K$ (i.e. the real K-dimensional Euclidean space) from sample i and, consequently, for P, N > K (which is always the case) the matrix \mathbf{R} should theoretically have rank K. This is simply the fundamental principle of dimension preservation from linear algebra. Of course, noise will make \mathbf{R} of maximum rank (min (N, P)) and in practice SVD-based methods are used to determine it's pseudorank.

It can be easily seen (as in the example of the MOS gas sensors above) that even if the theoretical model establishes the response variables R_j as linear expressions with respect to general univariable functions of each concentration, i.e.

$$R_{i}(c_{1},...,c_{K}) = A_{i1}f_{1}(c_{1}) + \dots + A_{iK}f_{K}(c_{K})$$
(8)

linear methods can still be used to determine the pseudorank of the data matrices and models can be constructed with respect to the 'abstract' concentrations $C_m = f_m(c_m)$.

Now, in a very general situation, if the response variables R_j are theoretically given by nonlinear functions of the kind

$$R_{i}(c_{1},...,c_{K}) = F_{i}(A_{j1}f_{1}(c_{1}) + \dots + A_{jK}f_{K}(c_{K}))$$
(9)

then univariable non-linear transformations of R_j (such as the inverse function of F_j) may restore the new response variables to the case previously described.

However, if any one sensor yields a response that is a non-linear function of all the concentrations, the dimension invariance principle of linear algebra cannot be applied in a simple way. The resulting row vectors of the matrix \mathbf{R} will not necessarily be close to a K-dimensional linear manifold because of the dimensional distortion effect caused by non-linearities.

Geometrically this can be better understood if K = 2 and P = 3 as in the ideal gas example mentioned above. In the linear case the points given by the N row vectors of **R** would be close to a plane in \mathbb{R}^3 (which can be determined by least squares). Now a non-linear function can be easily imagined which would fold this two-dimensional sheet in \mathbb{R}^3 in such a way that no plane would be a good approximation to those N points. In such a case there is no justification for the direct application of the elementary principle of dimension invariance from linear algebra.

The much more general topological principle of dimension invariance valid for smooth non-linear transformations¹⁹ cannot have a direct practical use here, since there is no simple non-linear SVD algorithm for the determination of the intrinsic (invariant) dimension. Actually, there is not much hope of designing a method which could handle all types of non-linear processes in a reasonable way and, to some extent, a restrictive hypothesis must be assumed in order to focus on appropriate techniques. The topological complexity that would be allowed in the pursuit of a general method is enormous and cannot be approached by a single path.¹²

In the present paper the main restriction is that a linearizing transformation T is sought only among functions which can be represented by a P-tuple of P (possibly different) univariable real functions, i.e.

$$T(x_1, \dots, x_p) = (T_1(x_1), \dots, T_p(x_p))$$
(10)

This restriction will simplify enormously the search for the appropriate T and will not be too severe on the scope of the method considering the vast applications of old function-specific methods (log linearization, Box-Cox, etc.) which are particular cases of it. Also, the Kolmogorov theorem on representation of real functions of several variables²⁰ suggests that this method might be applicable to very general situations if the functions $T_j(x)$ are appropriately constructed.

Thus the main goal is to design an algorithm which can construct an appropriate linearizing function of the above type for each non-linear process. The specific function when applied to the original response (vector) variable of the non-linear process will produce a new response (vector) variable which can be treated by the usual linear methods for rank determination and, eventually, model building. It is not always expected that the new response variables will have a direct linear relationship with the original components, but some important information, e.g. their number (rank determination), will certainly be revealed.

For example, if the function

$$R:\mathbb{R}^{K} \to \mathbb{R}^{P}, \quad R(c_{1}, c_{2}, \dots, c_{K}) = R(\mathbf{c}) = (R_{1}(\mathbf{c}), \dots, R_{P}(\mathbf{c}))$$
(11)

(theoretically) represents the responses of an ordered array of P sensors to a sample composed of K substances of concentrations c_1, \ldots, c_K , the main goal of the method is to look for a function

$$T: \mathbb{R}^{P} \to \mathbb{R}^{P}, \quad T(\mathbf{x}) = (T_{1}(\mathbf{x}), \dots, T_{P}(\mathbf{x}))$$
(12)

from the above class such that the process represented now by a theoretical model obtained by the composition of function T with model R (i.e. $T \circ R$) can be treated by linear methods. This requirement is obviously too vague, because the null function would be a trivial example of such T and, if the dimension (rank) is to be preserved, it is necessary to impose the condition that T is invertible. This is a very hard condition to characterize in the general case but, considering the restricted functional form assumed for T, will be attained simply by requiring that every coordinate function $T_i(x)$ must be strictly monotone. Let S be the set of T satisfying those conditions.

The general arguments for designing an algorithm to calculate an appropriate linearizing transformation T for a non-linear process R will rely on the topological principle of dimension invariance for smooth functions.¹⁹ Suppose that the non-linear function R has rank K. This means in practical terms that at least K of the P sensors are different and technically this can be represented by the rank of the Jacobian matrix

$$\frac{\partial R_j}{\partial c_i} \tag{13}$$

Then, given a cloud of points in \mathbb{R}^{K} (samples), the set of transformed points in \mathbb{R}^{P} via R (i.e. the data matrix of responses) will lie on a (non-linear) manifold of dimension K. When R is linear, they lie on a hyperplane of dimension K in \mathbb{R}^{P} , but in the general case this transformed cloud of points will certainly define a higher-dimensional hyperplane due to non-linear distortion. If the points are transformed again by any invertible function

$$T: \mathbb{R}^{P} \to \mathbb{R}^{P} \tag{14}$$

they will continue to lie on a (possibly different) manifold of (invariant) dimension K. However, the hyperplane defined by them can have a lower dimension than previously, the least possible dimension being K, and this is exactly the characterization which will be used to construct the linearization. The global linearizing transformation (GLT) T^* will be such that the data matrix

$$\mathbf{R}_{ii} = R_i(c_1, \dots, c_{Ki}), \quad \text{denoted by } \mathbf{R} = R(\mathbf{C})$$
(15)

is transformed into a new data matrix

$$\mathbf{Y}_{mn} = T_n(\mathbf{R}_{mn}), \quad \text{denoted by } \mathbf{Y} = T(\mathbf{R})$$
(16)

of smallest possible rank (K).

A numerical evaluation for this criterion can be given by

$$J\{T\} = \|\mathbf{Y} - \mathbf{Y}^K\| \tag{17}$$

where $\|\cdot\|$ is any matrix norm and \mathbf{Y}^{κ} is the K-dimensional truncation of matrix \mathbf{Y} obtained from singular value decomposition. The general algorithm for obtaining the global linearizing transformation T^* is to calculate the extremum problem

$$\min_{T \in S} J\{T\}$$
(18)

The main difficulty in formulating a practical algorithm for this problem is the fact that the set S is too large and cannot be described by a finite number of parameters. In order to remedy this situation, an appropriate set $\{I_j\}$ of linearly independent functions from S will be chosen in such a way that by linear combinations of the type

$$T_j(x, a_1, ..., a_n) = \sum_m a_m I_m(x)$$
 (19)

where $a_m \ge 0$ and $\sum a_m = 1$, the set S can be approximately described. As has already been pointed out, these functions can be chosen in many different ways. In the examples treated in this paper, a special basis of spline functions introduced by Ramsay¹⁸ has been used and their detailed construction is presented in the Appendix.

Assume for technical convenience that the raw data \mathbf{R}_{ij} are scaled between zero and one and call them \mathbf{X}_{ij} . Let us now call S the set of functions

$$T: [0, 1]^{P} \to [0, 1]^{P} \tag{20}$$

which can be represented in the form

$$T(x_1, \dots, x_P) = (T_1(x_1), \dots, T_P(x_P))$$
(21)

where each T_i : [0, 1] \rightarrow [0, 1] is continuous and strictly monotonic.

If the family is described parametrically by the coefficients $a_1, ..., a_n$ such that $\mathbf{Y}(\mathbf{X}, a_1, ..., a_n)$ are the general transformed data, all that must be done is to minimize the function

$$f(a_1, ..., a_n) = \| \mathbf{Y}(\mathbf{X}, a_1, ..., a_n) - \mathbf{Y}^K(\mathbf{X}, a_1, ..., a_n) \|$$
(22)

under the above-mentioned constraints (equation (19)) on a_1, \ldots, a_n , which can be accomplished with any of the existing optimization codes. The norm $\|\cdot\|$ can be chosen in a variety of ways as has already been pointed out. In this paper the Frobenius norm (which means a least squares approximation) was selected for the Taguchi sensor array example and a maximum likelihood approach for the ideal gas. If the 'hidden' dimension K is not known a priori, the above argument can still be used, but then Y^* must be chosen such that (17) is minimized also with respect to K.

The argument presented in this section has its origin in the work of Kruskal and Shepard,²¹ which was also later used by Ramsay.^{17,18}

In this paper M- and I-spline functions such as introduced by Winsberg and Ramsay¹⁸ were used to build the family of non-linear transformations and the optimizations were performed using MATLAB.

EXAMPLE: IDEAL GAS

This first example is presented as an illustration of the method. It was deliberately chosen in a simple and well-known context where the log function is the obvious linearizing transformation so that a comparison can be made with the constructed result following a tradition initiated by Kruskal and Shepard.²¹

A simulated data set was generated by the state equation for one mole of ideal gas (i.e. the molecules occupy no volume and have no interaction forces between them). For a given range of temperature (T) and pressure (P) the values of eight different properties (variables) were calculated for one mole of benzene. These properties are listed in Table 1. Note that these eight variables are defined as functions of temperature and pressure. The temperature values ranged from 300 to 1000 K in 70 K increments and for each temperature value ten different values of

Variable	Mathematical expression ^b	Units
Temperature	T	K
Pressure	Р	Pa
Volume	$V = \frac{RT}{P}$	m³
Isobaric coefficient of thermal expression	$\alpha = \frac{1}{T}$	K ⁻¹
Isothermal compressibility	$\beta = \frac{1}{P}$	Pa ⁻¹
Diffusion coefficient	$D = \frac{1}{3} \lambda \bar{c} = \frac{2}{3\sigma P} \left(\frac{kT^3}{m\pi}\right)^{1/2}$	$m^2 s^{-1}$
Coefficient of viscosity	$\eta = \frac{1}{3\sqrt{2}} \frac{m\bar{c}}{\sigma} = \frac{3}{2\sigma} \left(\frac{kTm}{\pi}\right)^{1/2}$	kg s ⁻¹ m ⁻¹
Coefficient of thermal conductivity	$\kappa = \frac{1}{3} \lambda \vec{c} \frac{C_{\rm v}}{V} = \frac{4}{\sigma} \left(\frac{k^3 T}{m\pi}\right)^{1/2}$	$J m^{-1} K^{-1} s^{-1}$

Table 1. Ideal gas variables

^a See e.g. Reference 22.

b R, ideal gas constant (8·3144 Jmol⁻¹K⁻¹); m, mass of one molecule of benzene (129·2983×10⁻²⁷ kg); k, Boltzmann constant (1·3807×10⁻²³ JK⁻¹); σ , benzene collision cross-section (0·88×10⁻¹⁸ m²); $\bar{\sigma}$, mean velocity of molecules, $\bar{c} = (8kT/m\pi)^{1/2}$; λ , mean free path, $\lambda = (1/\sqrt{2}) kT/5\sigma P$; $C_{\rm v}$, heat capacity at constant volume, computed from classical partition function (6*R*).

pressure were used from 0.15×10^6 to 1.5×10^6 Pa, equally spaced, generating a 110×8 data matrix **R**.

Six of the variables are non-linearly related to T and P, by means of products of their powers and of course the log function can be used to transform the values of each variable so that the redundancies can be expressed as a linear combination of the transformed variables log T and log P. In other words, there exists a set of eight analytical monotone transformations (actually the same log function) which make $\mathbf{Y} = T(\mathbf{R})$ approximately bidimensional (pseudorank two) and where the factor scores for each observation are log T and log P.

A normally distributed high level of random error (10%) was added to each variable. Figure 2 shows the plots of each variable versus the log function. Note that although the function is the



Figure 2. Ideal gas example: untransformed data R (with 10% error) versus log R for each variable defined in Table 1

same, it is plotted in different scales and ranges. Each variable of the raw data was then mean centered and scaled between zero and one, generating the matrix X as a normalizing procedure convenient for the application of the transformation. The transforming functions $T_j(x)$, $1 \le j \le P$, were made from order-two *M*-splines with two interior knots located at the tertiles for variables 1, 2, 4, 5 and 7. For variables 3, 6 and 8 one of them is at the first tertile and the other slightly to the right of the second tertile. By using these functions, the transformed matrix $Y^* = T^*(X)$ which has the closest matrix of rank two was obtained.

The estimated transformed data $\mathbf{Y}^* = T^*(\mathbf{X})$ of rank two are plotted in Figure 3 against the true transformed data log **R**. The linear relationship is shown to be better for some than for others, but quite visible for all the variables.



Figure 3. Ideal gas example: transformed data $Y^* = T^*(X)$ versus log **R** for each variable defined in Table 1. Order-two *M*-splines with two interior knots were used to obtain the transformation

Figure 4 shows the effect of the transformation in the dimensional structure of the data. It is clear that the transformed data $Y^* = T^*(X)$ have a sharp two-dimensional structure as opposed to that of the untransformed data matrix X; the first two singular values of $Y^* = T^*(X)$ are predominant against the remaining ones. Of course the log-transformed data log X also show a two-dimensional structure as expected. Actually it can be seen (Figure 4) that the log linearization and the GLT method give very close results for the singular value analysis.

Figure 5 displays the results obtained by the use of three interior knots for each variable, one located close to the inferior end, one in the middle and the third close to the superior end of the data, following a strategy suggested by Hastie and Tibshirani.⁹ Note that the use of this additional knot did not alter these transformations significantly.

It is important to analyze the behavior of the transformed data using a different dimensional structure. Dimensionalities one, two and three were used for the estimated transformation, maintaining the knot positions selected above. The results are shown in Figure 6, where the singular values (log scale) of the untransformed data X and transformed data $Y^* = T^*(X)$ are plotted for the three different cases. The transformation functions obtained for rank one (not shown) cannot linearize some of the variables. From these results it is clear that the data really have a bidimensional structure and this example shows how this procedure can be useful for determining the pseudorank of non-linear processes.

The same kind of analysis can be extended to non-ideal gases such as those which follow van der Waals' equation, with the difference that in this case explicit theoretical expressions are not available for comparison. Instead of extending this example to the van der Waals gases, a real data set which is known to be non-linear and for which a theoretical linearizing transformation is not available will be analyzed in the next section.



Figure 4. Ideal gas example: log of singular values versus number of factors before and after GLT and log transformations (mean-centered data)



Figure 5. Ideal gas example: transformed data $\mathbf{Y}^* = T^*(\mathbf{X})$ versus log **R** for each variable defined in Table 1. Order-two *M*-splines with three interior knots were used to obtain the transformation

APPLICATION: RANK DETERMINATION AND MODEL BUILDING FOR THE TAGUCHI SENSOR ARRAY DATA

Metal-oxide-semiconductor gas sensors were introduced by Taguchi in the early 1960s and since then have received much attention in the chemical literature. The signal originates from the interaction of the analyte and oxygen with the solid metal oxide surface. They have been used for multicomponent gas analysis.

However, their low-cost versions are non-selective and exhibit considerable crossinteractions, resulting in a non-linear behavior.²³ Consequently, it is desirable to develop methods which could help extract useful information from data given by a sensor array.



Figure 6. Ideal gas example: log of singular values versus number of factors for untransformed data (with 10% error) and transformed data for ranks one, two and three (mean-centered data). Second-order *M*-splines with one interior knot were used to obtain the transformation

Reliable theoretical formulae for the signal R_i produced by a sintered SiO₂ gas sensor as a function of gas concentration c_i were obtained²³ and can be written as

$$R_{j} = R_{0j} \left(1 + \sum_{i=1}^{K} A_{ji}(c_{i})^{m_{ij}} \right)^{-\beta_{j}}, \quad 1 \le j \le P$$
(23)

where A_{ji} and m_{ij} are parameters depending upon both the gas and the sensor and R_{oj} and $\beta_j > 0$ are only dependent upon the sensor. The general problem from a chemometric point of view is to estimate concentrations given the instrument output \mathbf{R}_{ij} . Of course it is clearly impossible to produce an analytic inverse for the above formulae even for the case of P = K. As in the linear case, the main goal is to develop a method which will give the desired information with an 'excess' (redundant) of output, i.e. when $P \gg K$.

The data set used in this application was collected by W. P. Carey from the Department of Electrical Engineering, University of Washington, using an array of eight Taguchi gas sensors which are non-linear in their response characteristics.²⁴ The raw data set contains the response from 20 different mixtures of organic solvent vapors of toluene and benzene, replicated five times with concentrations varying from 5 to 500 ppm, generating a 100×8 data matrix. This data set has been analyzed using various linear and non-linear methods for calibration by Sekulic *et al.*²⁵ where, from 100 samples, 50 were used as a calibration set to build the model and 50 for prediction.

The first step in the application of the GLT method to this problem will be to obtain the best linear linearizing transformation T^* , (equation (18)) of a given dimensional structure by using spline functions and the Frobenius norm. The data matrix **R** is the 50×8 calibration set used by Sekulic *et al.*²⁵

The raw data had values between zero and one and did not require scaling. Mean centering was applied to each variable to give the X-matrix. One knot per variable was used with second-order *M*-splines.

The main result obtained from this first step is the determination of the correct rank of the data. The spline transformation was applied with predefined ranks *two* and *three*. The locations of the knots were optimized independently for each variable and the best linear combination of the spline basis set (best transformation functions) which reduced the dimensional structure from P = 8 to K = 2 or 3 was determined. The results are compared with the untransformed data in Figure 7. The difference between the second and third eigenvalues is greatest when a rank-two model is used for the transformation. It is obvious then that the transformation made the calibration set clearly bidimensional. In the same figure it is shown that with a linear direct procedure the process seems to be full rank.

The second step, which is a much more difficult problem, is the use of the linearized transformed data matrix $\mathbf{Y}^* = T^*(\mathbf{X})$ to construct models for the concentrations of toluene and benzene by the well-known PCR method.^{3,4} The final step will be to estimate the concentrations of the prediction set samples.

Once the rank of the system has been identified, the calibration model for this specific rank is constructed. Since the calibration and prediction sets are available, the root mean square error of prediction given by

RMSEP =
$$\left(\frac{\sum_{i=1}^{N} (c_i - \hat{c}_i)^2}{N}\right)^{1/2}$$
 (24)

can be used to select the appropriate model. Here c_i is the concentration of sample *i* from the prediction set and \hat{c}_i is the corresponding value estimated by the calibration model for the *i*th



Figure 7. Taguchi sensors example: log of singular values versus number of factors for untransformed data and transformed data for ranks two and three (mean-centered data). Second-order *M*-splines with one interior knot were used to obtain the transformation

sample. N is the number of samples in the prediction data set. As suggested by Sekulic *et al.*,²⁵ the results are expressed in terms of the percent relative prediction given by

$$\% \text{REL.RMSEP} = \frac{\text{RMSEP} \times 100}{\bar{c}}$$
(25)

where \bar{c} is the mean concentration value in the prediction set of samples.

The model depends upon the knot selection (number and location) and the order of the *M*-splines, which has been chosen in this case to be two. Using one knot per variable, their locations were chosen to be those which yield the lowest %REL.RMSEP for the calibration set with bidimensional structure. The results are given in Table 2. Note that the linearizing transformation function of rank two gives results which improve upon the 'best' results obtained by PCR models with a much larger number of components to span the two-dimensional concentration space (shown in parentheses²⁵). Table 2 also contains the results given by direct application of the PCR method with the correct rank two, which shows the %REL.RMSEP above 48%.

Two knots for each variable were also employed and their locations were optimized for each variable at a time, considering a bidimensional structure model. As before, the least squares approach was used. The %REL.RMSEP is also given in Table 2. The results are slightly better than those for the previous case with only one knot per variable.

In order to determine how the transformation affected the original data, the transformed data $\mathbf{Y}^* = T^*(\mathbf{R})$ are plotted versus the raw data \mathbf{R} for each variable in Figure 8. Note that for variables 1, 4, 6 and 8, except at the lower end, the plots are basically linear. They are also all very close to each other, indicating that these variables are fairly proportional to the concentrations and certainly originated from very similar sensors. Actually the array consisted of only five different types of sensors.²⁴ From Figure 8 it can be seen that three of them are of the same type (4, 6 and 8), one gives a similar response (1) and also two others are of the same type (2 and 7). However, the most interesting result in Figure 8 is the extent of the transformation for variable 3 which from the plot can be considered to be the only one that is 'highly' non-linear. All the other variables show very mild non-linearity compared with variable

	Untransform	Untransformed data R ^a	
	PCR	PCR	
Toluene Benzene	22.57 (7) 22.11 (8)	51.68 (2) 48.66 (2)	
	Transformed data $\mathbf{Y}0 = T^*(\mathbf{R})$		
	One interior knot PCR	Two interior knots PCR	
Toluene Benzene	20·93 21·81	18·70 17·28	

Table 2. Percent relative prediction error (%REL.RMSEP) for Taguchi untransformed data and transformed data of rank two using one and two interior knots

"Value in parentheses refer to number of factors incorporated in model.

3. Thus some improvement in the %REL.RMSEP might be expected if this sensor (variable) were excluded from the array. These results are shown in Figure 9, where the %REL.RMSEP values obtained from the PCR models for the raw data **R** are compared, first taking into account and then ignoring the results from the third sensor. The symbols \bigcirc indicate the %REL.RMSEP obtained by the GLT method with rank two (from Table 2) and the symbols ^{*} indicate the number of factors chosen in Reference 25 to build the calibration model. Upon removing the third variable, the shape of the curve for the standard error of prediction versus the number of factors in the model changes drastically for the PCR methods. There is a decrease in the %REL.RMSEP of approximately 19% (from 55 to 36) and 24% (from 48.5 to 24.5) for toluene and benzene respectively when the number of factors in the PCR models is increased from one to two. When variable 3 is included in the model, this decrease is only 4.5% (from 56.2 to 51.7) for toluene and no decrease in the %REL.RMSEP is observed for benzene. The benzene plot in Figure 9 shows almost no decrease in the prediction error for models with more than two factors. Thus sensor 3 is responsible for a high degree of non-linear behavior in the data set.



Figure 8. Taguchi sensors example: untransformed data versus transformed data for each variable. Second-order *M*-splines with two interior knots were used to obtain the transformation



Figure 9. %REL.RMSEP using PCR models for all sensors (------) and after removing sensor 3 (-----): O, %REL.RMSEP obtained by GLT method of rank two (using two interior knots); *, number of factors used in Reference 25 to build calibration model

Third-order piecewise polynomials were considered and, as in the case of the ideal gas, no significant changes were observed in the results.

This example clearly shows the exploratory data analysis advantages derived from this method. Besides being useful for determining the true effective rank of a data set, the method can uncover non-linearities which can be avoided when linear models are employed or transformed to improve the model-building process. In the present case it became obvious (Figure 8) that variable (sensor) 3 is the main and almost the only source of high non-linearity. It is shown how this procedure can lead to a rank-two model (the true rank) with prediction accuracy better than that obtained by linear models of excessive rank.

For both examples studied in this paper, the knot selection for exploratory data analysis did not require any exhaustive knot optimization. They were found heuristically by a slight displacement from their initial position (median, tertiles) in directions which improve equation (17). The model building is more sensitive to the knot positions, but, on the other hand, that could be found automatically if the knot positions were included as parameters in the optimization process.

CONCLUSIONS

It is shown in this paper that the difficult problem of rank determination with non-linear data can be successfully approached in many cases by the GLT method, which makes it a potential method for preprocessing data. The method is also shown to be efficient in uncovering nonlinearities. For those processes which are described by more general functional relations, the method gives a global linearizing transformation which can be used for model building by conventional linear methods.

Both examples treated have a low number of variables (eight), but the method can also be applied to a much larger number of variables, which is the case for spectral data where P = 1000 variables are common. In this case one can select a representative small number P_o of variables and apply the above method to obtain their P_o spline transformations. The whole data set can then be transformed, each variable by its respective representative transformation.

The theoretical core of this paper is the linearization scheme, which is formulated in such a way that it can be implemented by many different techniques depending on the class of functions $\{I_i\}$ and the matrix norm. This flexibility allows its use for a much broader range of non-linear processes.

The method of global linearization presented here is a powerful technique for rank determination that extends log linearization (and other function specific methods) to much more general situations. Of course it is not intended for problems where linearization is not necessary or where it can be achieved with known elementary functions. On the other hand, like any useful non-linear method, it cannot be completely general for reasons of complexity. Instead, its range of applications covers an important middle ground between the familiar and the unattainable extremes and it has the advantage of conceptual and computational simplicity and vast possibility of generalization.

One might suspect that for some highly non-linear problems (i.e. where there are 'strong interactions' between independent variables) even more general kinds of transformations would be necessary instead of the univariable functions employed here. However, at least from a theoretical point of view, Kolmogorov's approximation theory ensures that univariable functions are sufficient for the most general linearization if used appropriately. Understandably, its practical implementation will make the problem vastly more complex and will be left for future work where more sophisticated topological and analytical methods are to be employed.

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APPENDIX: THE MONOTONE SPLINES

Spline functions have their origin in the problem of describing an interpolating curve through a given set of points under the condition of minimum elastic energy. Cubic splines, for example, are defined by the minimum for the functional

$$J(F) = [y_i - F(x_i)]^2 + \lambda \int [F'(s)]^2 ds$$
(26)

among twice continuously differentiable functions and are constructed analytically as piecewise polynomials. The integral term imposes a penalization for wiggling (with weight $\lambda > 0$) and accounts for a balance between its interpolating character and its smoothness.²⁶ Their theory has been extensively developed together with applications to many different areas^{9,16,26,27} since the late 1970s.

In this paper, splines of a special type introduced by Ramsay¹⁷ (M- and I-splines) are used as functional blocks for the approximate description of the class of functions S.

M-splines of order *k* on a closed interval [a, b] are constructed as piecewise polynomials of degree k-1 in a mesh of previously determined *q* subintervals described by a sequence of junction points $\{\xi_j\}$, where $a = \xi_1 < \cdots < \xi_{q+1} = b$. Within each subinterval $[\xi_j, \xi_{j+1}]$ the function is represented by a polynomial of order k-1 and by matching smoothness conditions at the boundaries. For the case where continuity is maximum, two polynomials from adjacent intervals are required to have equal derivatives or all orders from zero to k-2 at a common point. Given the order *k* and the degree of smoothness required, the spline functions are usually defined by a sequence of numbers called knots, $\{t_i\}$, as described in the following. One single knot is placed at each interior junction point ξ_j and *k* knots are placed at each of the endpoints *a* and *b*.

The initial total number of degrees of freedom is kq, i.e. the number of parameters necessary to characterize a polynomial of degree k-1 times the number q of subintervals. Now, if the matching conditions (k-1) for each interior point) are deducted, it is concluded that the dimension of the function space associated with the *M*-splines is

$$n = kq - (k-1)(q-1) = k + q - 1$$
(27)

Thus for a given selection of order and knots a family of M_i -splines (set of basis functions) is defined recursively¹⁷ by

$$M_{i}(x, 1, t) = \begin{cases} 1/(t_{i+1} - t_{1}) & \text{for } x \in [t_{i}, t_{i+1}) \\ 0 & \text{otherwise} \end{cases}$$

$$M_{i}(x, k, t) = k \left((x - t_{i})M_{i}(x, k - 1, t) + \frac{(t_{i+k} - x)}{(k - 1)(t_{i+k} - t_{i})} M_{i+1}(x, k - 1, t) \right) \quad \text{for } k > 1$$

$$(28)$$

The interesting properties of *M*-splines are that they are positive in each interval $[t_i, t_{i-1})$ and zero elsewhere and also normalized,

$$\int_{a}^{b} M_i \,\mathrm{d}x = 1 \tag{29}$$

The construction of *I*-splines, which are actually the ones used in the paper, is essentially achieved by integration of the *M*-splines. For $x \in [t_i, t_{i+1})$ they can be defined by

$$I_{i}(x,k,t) = \begin{cases} 0 & \text{for } j < i \\ \sum_{m=i}^{m=j} \frac{t_{m+k+1} - t_{m}}{k-1} M_{m}(x,k+1,t) & \text{for } j-k+1 \le i \le j \\ 1 & \text{for } i < j-k+1 \end{cases}$$
(30)

From the non-negativity of *M*-splines the monotonicity of *I* is assured and it must also be noted that $I_i(a) = 0$ and $I_i(b) = 1$.

Once the basis set has been defined as above, the construction of a family of monotone functions $T_j(x, a_1, ..., a_n)$ is carried out by linear combination under the restriction that the coefficients must be non-negative and have sum equal to one, i.e.

$$T_{j}(x, a_{i}, ..., a_{n}) = \sum_{r=1}^{r=n} a_{r} I_{r}(x)$$

$$a_{r} \ge 0$$

$$\sum_{1}^{n} a_{r} = 1$$
(31)

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