Use of Transformations on Parameters in Non-Linear Theory. II.
Measures of Non-Linearity and Transformations to Reduce
Non-Linearity

by

Irwin Guttman and Duane A. Meeter

1 This research was supported by the Office of Naval Research under NONR 1202 (17), the Wisconsin Alumni Research Foundation and the National Science Foundation.

2 Now at the Department of Statistics, The Florida State University, Tallahassee, Florida.
Introduction

In the first of these two papers, the authors (1964) have discussed the use of transformations on parameters designed to accelerate convergence in non-linear least squares problems. In this paper, we continue our discussion of non-linear least squares problems, and turn attention to measures of non-linearity.

In Section 1, we introduce some measures of non-linearity due to Beale (1960 a), and in Section 2, we examine the behavior of these measures using certain specific examples. Finally, in Section 3, we investigate the effect of using transformations of parameters to reduce the non-linearity measures.

We will refer to the first paper [Guttman and Meeter (1964)] in the sequel as I.

1. Some Measures of Non-Linearity

As is well known, the results concerning the precision of the least-squares estimates and the adequacy of the model, when that model is linear in the parameters, are only approximately true in the non-linear case. It is reasonable, then, to develop some measure of non-linearity which would indicate when we are justified in using the linear theory results as approximations for non-linear models, and possibly indicate how the linear theory results could be modified to be better approximations. Some such measures have been proposed by Beale (1960 a), and we now examine the validity of these measures.
Suppose then, we again consider the model

$$
\eta = f(\theta, \xi)
$$

(1.1)

where $\theta$ is a (p x 1) vector of parameters and $\xi$ is a vector of independent variables. We suppose that $n$ independent observations on the response $\eta$ have been made, say $(y_1, \ldots, y_u, \ldots, y_n)$, where the $u$-th observation $y_u$ was obtained when the independent variables $\xi$ were set at $\xi_u$. Of course, we are concerned here with the case where $\theta$ enters the model (1.1) non-linearly.

If $\hat{\theta}$ denotes the least squares estimates of $\theta$, the tangent plane approximation to the solution locus—see I—in the neighborhood of $\hat{\theta}$ is given by

$$
\tau_u(\theta) = \eta_u(\hat{\theta}) + \sum_{i=1}^{p} (\theta_i - \hat{\theta}_i) \left. \frac{\partial f(\theta, \xi_u)}{\partial \theta_i} \right|_{\theta = \hat{\theta}}, \ u=1, \ldots, n,
$$

(1.2)

or

$$
\tau(\theta) = \eta(\hat{\theta}) + X(\theta - \hat{\theta}),
$$

and where $\eta_u(\hat{\theta}) = f(\hat{\theta}, \xi_u)$. Of course, $\tau(\theta)$ differs from the actual point $\eta(\theta)$ which is on the solution locus, because of the non-linearity of (1.1). Suppose we consider points $\hat{\theta}_w$, $w = 1, \ldots, m$, in the neighborhood of $\hat{\theta}$; then a crude measure of non-linearity would be
\[ Q_\theta = \sum_{w=1}^{m} \sum_{u=1}^{n} \left( \eta_u (\theta_w) - \eta_u (\hat{\theta}) \right)^2 = \sum_{w=1}^{m} \left( \eta_w (\theta) - \eta_w (\hat{\theta}) \right)^2 \]  

(1.3)

\[ Q_\theta \] is the sum of squares of the distances (in sample space) from the points \( \eta(\theta_w) \) to the associated points \( \tau(\theta_w) \) on the tangent plane. As explained in Beale (1960 a), to normalize this measure \( Q_\theta \) is divided by the quantity

\[ \sum_{w=1}^{m} \left( \eta_u (\theta_w) - \eta_u (\hat{\theta}) \right)^2 = \sum_{w=1}^{m} \left( \eta(\theta_w) - \eta(\hat{\theta}) \right)^4 \]  

(1.4)

Since \( Q_\theta \) has the dimension of the square of an observation, and (1.4) the dimension of the fourth power of an observation, the quantity

\[ \hat{N}_\theta = p s^2 \sum_{w=1}^{m} \left( \eta(\theta) - \tau(\theta_w) \right)^2 / \sum_{w=1}^{m} \left( \eta(\theta) - \eta(\hat{\theta}) \right)^4 \]  

(1.5)

where \( s^2 \) is our best estimate of \( \sigma^2 \) (the variance of the observations), is a dimensionless quantity that can be regarded as the estimated normalized measure of the non-linearity of the model when expressed in terms of the parameters \( \theta \). The reason for the factor \( p \) will be explained later. Beale states that the value of \( \hat{N}_\theta \) may depend on the configuration of the points \( \eta(\theta_w) \) around \( \eta(\hat{\theta}) \), but should not depend significantly on the number \( m \) of points used, or their distances.
from $n(\hat{\theta})$, if these distances are not too great.

Corresponding to the empirical measure of non-linearity $\hat{N}_\theta$
given by (1.5), there is a theoretical measure of non-linearity, $N_\theta$.
It is formed from $\hat{N}_\theta$ by replacing $s^2$ by $\sigma^2$, and letting the values of
$\theta_w$ be an infinite set of values of $\theta$ such that the points $\tau(\theta_w)$ have
a p-dimensional spherical normal distribution about $n(\hat{\theta})$ with arbitrarily
small variance. Both $N_\theta$ and $\hat{N}_\theta$ are invariant under any linear
transformation of parameters, (in the case of $\hat{N}_\theta$, the values $\theta_w$, $w=1, \ldots,
m$ must be held constant,) and also invariant under any orthogonal linear
transformation of coordinates in sample space.

Now suppose we made arbitrary transformations of parameters,
say $\varphi = \varphi(\theta)$. Suppose the minimum value of $N_\theta$ under these trans-
formations is attained by using the transformation $\hat{\Phi} = \hat{\varphi}(\theta)$, and is denoted
by $N_\phi$. Then the geometrical interpretation of $N_\phi$ is that it is the value
of $N_\theta$ when the parameters are transformed in such a way that $\tau(\theta)$ is
always at the foot of the perpendicular from $n(\theta)$ to the plane tangent
to the solution locus at $n(\hat{\theta})$. Equivalently, $N_\phi$ is the minimum possible
value of $N_\theta$ if the model and the experimental design are fixed but any
transformation of parameters is permitted -- $N_\phi$ can be called the intrinsic
non-linearity of the model. A detailed description of how $N_\theta$ and $N_\phi$,
the theoretical measures of non-linearity, can be estimated in practice
is given by Beale (1960 b).
(We remark that by minimizing $N(h)$, where $N(h)$ is given by (1.26) of I, we were minimizing (apart from a factor $ps^2$) the
umerator of Beale's empirical measure of non-linearity, $\hat{N}_\theta$, while holding the denominator constant. However, we were minimizing $\hat{N}_\theta$ within the class of transformations defined by (1.16) of I, and in the neighborhood of $\hat{\theta}^{(r)}$, the $r$th approximation to $\hat{\theta}$.)

There is a minimum value $\hat{N}_\phi$ of the empirical measure of non-linearity $\hat{N}_\theta$, corresponding to $N_\phi$ and $N_\theta$. Suppose that the transformation $\phi = \phi(\theta)$ is such that

$$
\phi(\hat{\theta}) = 0, \quad \left. \frac{\partial \phi_i}{\partial \theta_j} \right|_{\hat{\theta}} = 1, \quad \left. \frac{\partial \phi_i}{\partial \theta_j} \right|_{\hat{\theta}} = 0, \quad j \neq i.
$$

(1.6)

(This can always be achieved by applying a final, linear transformation to a given transformation, without affecting the value of $\hat{N}_\phi$.) $\hat{N}_\phi$ is computed by writing

$$
Q_\phi = \sum_{w=1}^m \sum_{u=1}^n \left[ \eta_u (\hat{\theta}_w) - \eta_u (\hat{\theta}) - \sum_{j=1}^p \phi_{w,j} \frac{\partial f(\theta, \xi_u)}{\partial \theta_j} \right] (\hat{\theta})^2
$$

where the $\phi_{w,j}$ are chosen to minimize $Q_\theta$; they are obtained from the normal equations

$$
X^\top [ \eta(\theta_w) - \eta(\theta) - X \phi_w ] = 0, \quad w = 1, \ldots, m, \quad (1.7)
$$
which define \( m \) sets of \( p \) simultaneous linear equations. Then the value of \( N_\phi \) which corresponds to \((1.5)\) is given by

\[
N_\phi = ps^2 Q / \sum_{w=1}^{m} || \eta(\theta_w) - \eta(\hat{\theta}) ||^4.
\]

(1.8)

This measure of non-linearity is now used to allow for the effect of non-linearity on the usual linear theory results as follows. Suppose that we wished to use as a confidence region for a non-linear model the values of \( \theta \) for which

\[
S(\theta) - S(\hat{\theta}) \leq ps^2 F_{p, \nu; \alpha},
\]

(1.9)

where \( S(\theta) \) is the sum of squares \( \sum_{u=1}^{n} [ y_u - f(\theta, \xi_u) ]^2 \), \( s^2 \) is an independent estimate of \( \sigma^2 \) with \( \nu \) degrees of freedom, and \( F_{p, \nu; \alpha} \) is the upper \( 100\alpha\% \) point of the \( F \) distribution with \( (p, \nu) \) degrees of freedom. Beale shows that, if \( N_\phi \) is not too large, \((1.9)\) is a confidence region with associated probability greater than or equal to \( 1-\alpha \) if the right hand side of \((1.9)\) is multiplied by

\[
1 + N_\phi \quad (p=1)
\]

\[
1 + \left( \frac{p + 2}{p} \right) N_\phi \quad (p \geq 2).
\]

(1.10)

*We are assuming that the errors in the observations \( y_u \) are independent \( N(0, \sigma^2) \) random variables.*
Multipliers similar to (1.10) have also been given by Beale (1960 a) for the following cases:

(i) \( S_{\hat{\theta}}^2 / n-p \) replaces \( s^2 \) in the right hand side of (1.9).

(ii) A lack of fit test employing the inequality

\[
S_{\hat{\theta}}^2 \leq (n-p)s^2 F_{n-p, \nu; \alpha} \quad (1.11)
\]

To gain an idea of the significance of particular values of the non-linearity measures, Beale begins with the case when the model is essentially linear in \( \theta \). From (1.9) and usual linear theory results, a point on the boundary of this confidence region is obtained by moving (in parameter space) from \( \hat{\theta} \) to some point \( \theta^* \), where \( \theta^* \) is such that the point \( \tau(\theta^*) \) (obtained by linear extrapolation from \( \eta(\hat{\theta}) \) as in (1.2)) is at a distance

\[
d = \left( p s^2 F_{p, \nu; \alpha} \right)^{\frac{1}{2}} \quad (1.12)
\]

from \( \eta(\hat{\theta}) \) in sample space. If the model is truly linear in \( \theta \), the distance from \( \tau(\theta^*) \) to \( \eta(\theta^*) \), say \( \epsilon d \), will always be zero. Otherwise, \( \epsilon \) will not be zero, but if the points \( \eta(\theta_w) \) used in computing \( \hat{N}_\theta \) give a reasonable representation of the shape of the surface at
relevant distances from $\hat{n}(\bar{g})$, then

$$E(\epsilon d)^2 \overset{\sim}{=} \hat{N}_\theta d^4/ps^2.$$  \hfill (1.13)

Then, using (1.12),

$$E(\epsilon^2) \overset{\sim}{=} \hat{N}_\theta d^2/ps^2 = \hat{N}_\theta p_{\nu,\alpha}$$  \hfill (1.14)

*An argument leading up to (1.13) can be illustrated thus:

Since $||\hat{n}(\bar{g}) - \hat{n}(\bar{g})||^4 = d^4 + (\epsilon d)^4 - 2\epsilon d^4 \cos \alpha$, if $\epsilon$ is somewhat less than 1 we can approximate each of the terms $||\hat{n}(\theta_w) - \hat{n}(\bar{g})||^4$ in the denominator of $\hat{N}_\theta$ (see (1.3)) by $d^4$, which leads to (1.13).
Thus, Boole says that the model could be regarded as being
disastrously non-linear in $\theta$ if

$$\hat{N}_0 > \frac{1}{F_{p, \nu}},$$

(1.15)
since the root mean square value of the discrepancy vector
$\hat{\eta}(\hat{\theta}^*) - \hat{\eta}(\hat{\theta}^*)$ is then longer than the intended vector $\eta(\theta^*) - \eta(\theta)$.

One could also say that the linear approximation is
satisfactory if

$$\hat{N}_0 < 0.01/F_{p, \nu},$$

(1.16)
since the root mean square value of the discrepancy vector is less
than one-tenth of the length of the intended vector. (The primary
reason for inserting a factor $p$ in the definition (1.5) of $\hat{N}_0$ is to
avoid having a factor $p$ in the two previous inequalities.)

As $p$, the number of parameters, increases it becomes more and
more difficult to compute and appreciate the shape and significance
of a confidence region such as (1.9). In such cases it is desirable
(under the assumption that the observations $y_u$ are independently
normal with mean $\eta_u$ and variance $\sigma^2$) to be able to summarize the
information about the parameters by saying that $\hat{\theta} - \theta$ is approxi-
mately normally distributed with covariance matrix

$$(X'X)^{-1}\sigma^2, \text{ i.e., } \eta = \eta(\theta) + X(\theta - \hat{\theta}).$$

If this approximation
is valid, the linear theory confidence region

\[(\theta - \hat{\theta})' X' X (\theta - \hat{\theta}) \leq p \frac{S(\hat{\theta})}{n-p} F_{p, n-p; \alpha} \quad (1.17)\]

or a similar expression corresponding to (1.9), should produce values \(\hat{\theta}^*\) on its boundary such that actual sums of squares \(S(\hat{\theta}^*)\) would not be too different from the linear theory sum of squares

\[S(\hat{\theta}) = S(\hat{\theta}) + (\theta - \hat{\theta})' X' X (\theta - \hat{\theta}) \quad (1.18)\]

predicted for the boundary of (1.17).

Beale suggests that if \(\hat{N}_\theta\) falls between the levels stated in (1.15) and (1.16), the above approximations to the distribution of \(\hat{\theta} - \theta\) can give a rough picture of the significance of the experiment, but more analysis is needed to bring out the full implications of the data. One technique that might help is a (non-linear) transformation of parameters. Of course, it will not be possible to reduce \(\hat{N}_\phi\) below \(\hat{N}_\phi\), so if \(\hat{N}_\theta - \hat{N}_\phi\) is small compared with \(\frac{1}{F_{p, \nu; \alpha}}\), transformations will be of little value. If \(\hat{N}_\phi\) exceeds say \(0.1 F_{p, \nu; \alpha}\), then no transformation can reduce the non-linearity to negligible proportions. The covariance matrix for the transformed parameters will depend not only on \(\hat{\theta}\) but on the observed sample point \(x\).

Since \(\hat{N}_\phi\) is an estimate of \(N_\phi\), the approximate confidence regions of the type (1.9) are suspect if \(\hat{N}_\phi\) is as large as 1, but they are quite acceptable if \(\hat{N}_\phi\) is less than say 0.1, particularly if modified as suggested by (1.10).

For a number of reasons, Beale's non-linearity measures as described in the previous section are very appealing. Of course, as any good measures of non-linearity should, they take into account the non-linearity of the model itself. More importantly, the measures are also affected by the actual design of the experiment (i.e., the levels \( \xi_u \)) through the model \( \eta_u = f(0, \xi_u) \), and also by the size of \( \sigma^2 \), the variance of the observations \( y_u \). Further, they retain the likelihood contours as a basis for an explanation of the joint significance of the parameter estimates. In this section we will study with the aid of examples, the applicability of Beale's measures to actual situations.

The examples chosen arise out of a chemical system in which a substance A decomposes at a rate proportional to \( \theta_1 \) to form B which decomposes at a rate proportional to \( \theta_2 \) to form substance C. If \( \eta_1 \), \( \eta_2 \) and \( \eta_3 \) represent the respective yields of A, B, and C at time \( \xi \), under certain well-defined assumptions the simultaneous differential equations
\[
\frac{d\eta_1}{d\xi} = -\theta_1 \eta_1 \\
\frac{d\eta_2}{d\xi} = \theta_1 \eta_1 - \theta_2 \eta_2 \\
\frac{d\eta_3}{d\xi} = \theta_2 \eta_2
\] (2.1)

can be derived, with boundary conditions \(\eta_1 = 1, \ \eta_2 = \eta_3 = 0\) at time \(\xi = 0\), and where \(\theta_1, \theta_2 > 0\).

In this particular case the differential equations can be solved explicitly to obtain, for example, the yield of \(B\) at any intermediate time \(\xi\):

\[
\eta_2 = \frac{1}{\theta_1 - \theta_2} (\theta_1 e^{-\theta_2 \xi} - \theta_1 e^{-\theta_1 \xi})
\] (2.2)

and the corresponding yield of \(C\):

\[
\eta_3 = 1 - \frac{1}{\theta_1 - \theta_2} (\theta_1 e^{-\theta_2 \xi} - \theta_2 e^{-\theta_1 \xi})
\] (2.3)

where, because \(\theta_1, \theta_2 > 0, \ \ 0 \leq \xi < \infty\), we have \(0 < \eta_2, \eta_3 < 1\).

It turns out that for the levels of \(\theta\) chosen, \((\theta_1 = 1.4, \ \theta_2 = .4)\), these two models, so mathematically similar, represent fairly well the extremes of low and high non-linearity. In our examples, the basic design used for the model \(\eta_2\) was

\[
\xi = (0.25, 0.5, 1, 1.5, 2, 4)
\] (2.4)

while for \(\eta_3\) it was

\[
\xi = (1, 2, 3, 4, 5, 6)
\] (2.5)
In order to be able to analyze the effect of $n$, the number of
observations, the analyses were carried out with 4, 6, and 12 observa-
tions. For $\eta_2$, the four-point design was the subset

$$\xi = \{0.5, 1, 2, 4\}$$

of \{2,4\}, while for $\eta_3$ it was

$$\xi = \{0.1, 2, 4, 6\}.$$ 

The designs for $n = 12$ were obtained by replicating the designs \{2,4\}
and \{2,5\}.

The observations were constructed by adding random normal
deviates with standard deviation 0.025 to the true response. The same
12 normal deviates were used for the corresponding points $\eta_{2u}$ and
$\eta_{3u}$, $u = 1, \ldots, n$. The curves and the sample points are displayed in
Figures 2.1 and 2.2.

To obtain some idea of the extent of the non-linearity in the various
examples, contours of $S(\theta)$ were plotted. In each case, the level of
$S(\theta)$ plotted was obtained from the usual $100(1 - \alpha)\%$ linear theory
confidence region when $S(\hat{\theta})/(n-p)$ is used to estimate $\sigma^2$, viz.,

$$S(\theta) - S(\hat{\theta}) = \frac{S(\hat{\theta}) F_{p, n-p; \alpha}}{(n-p)}$$

with $\alpha = 0.05$. Figures 2.3 through 2.6 display the results for
the models $\eta_2$ and $\eta_3$ with 4, 6, and 12 observations. In each
figure, the upper drawing shows the actual $S(\theta)$ contour; the middle
drawing displays the contour obtained by assuming that the model is
Figure 2.1 The "true" model $\eta_2$ and the twelve observations.

Figure 2.2 The true model $\eta_3$ and the twelve observations.
a. Exact contour.

b. Contour from linear approximation. The points on the ellipse were used for the calculation of non-linearity measures.

c. Contour from quadratic approximation.

Figure 2.3 Contours for the model $\eta_3$; four observations.
a. Exact contour.

b. Contour from linear approximation. The points on the ellipse were used for the calculation of non-linearity measures.

c. Contour from quadratic approximation.

Figure 2.4 Contours for the model $\eta_2$; four observations.
a. Exact contours. The elliptical contour pertains to $\eta_2$.

b. Contour from linear approximation. The points on the ellipse were used in the calculation of non-linearity measures.

c. Contour from quadratic approximation

Figure 2.5 Contours for the models $\eta_2$ and $\eta_3$; six observations.
a. Exact contours. The elliptical contour pertains to $\eta_2$.

b. Contour from linear approximation. The points on the ellipse were used in the calculation of non-linearity measures.

c. Contour from quadratic approximation.

Figure 2.6 Contours for the models $\eta_2$ and $\eta_3$; twelve observations.
given by a linear expansion about \( \hat{0} \), and thus the contour is an ellipse; the lower drawing shows the contour we obtain when we assume that \( \eta(\theta, \xi) \) is given by a Taylor series expansion about \( \hat{0} \) including the second derivatives \( \frac{\partial^2 \eta}{\partial \theta_1 \partial \theta_2} \), i.e., \( \eta \) is given by an expression quadratic in the parameters.

Figures 2.3 and 2.4 demonstrate the great contrast between the model \( \eta_1 \) and \( \eta_2 \) with just four observations. The contour for \( \eta_2 \) lies well within the plotted region (the region is a rectangle whose sides are, approximately, of length two times the actual parameter values.) The \( \eta_3 \) contour is not closed within the plotted region, and cannot in any sense be said to be approximated by the elliptical contour, whereas the contour \( \eta_2 \) is reasonably well approximated by the elliptical contour, and very well approximated by the contours based on the second derivative expansion of \( \eta_2 \). Figures 2.5 and 2.6 show the results for 6 and 12 observations, respectively. The contour for \( \eta_2 \) in each case is superimposed on the \( \eta_3 \) contour for contrast; the discrepancies for the \( \eta_2 \) model contours between the exact, linear, and quadratic representations of the model are too small to be plotted on the given scale.

We observe that, at least for the case of 12 observations, the contours for \( \eta_2 \) and \( \eta_3 \) differ mainly in the amount of variation "assigned to the" parameter \( \theta_1 \). This phenomenon is readily understood when we note
the small discrepancy between the curves $\eta_1 (\theta_1 = 1.4, \theta_2 = .4, \xi)$ and $\eta_3 (\theta_1 \to \infty, \theta_2 = .4, \xi)$, displayed in Figure 2.7. Since $S(\theta)$ measures the sum of squared distances between the observations and the curve $\eta(\theta_1, \theta_2, \xi)$ at selected points along the curve, it is not surprising that $S(\theta)$ is insensitive to large changes in $\theta_1$. The curve $\eta_2 (\theta_2, \xi)$, on the other hand, can change shape considerably for relatively small changes in $\theta_1$ and $\theta_2$, and this, in part, accounts for its behaviour being more nearly linear in the neighborhood of $\theta$.

All of the points $\theta_w$, $w = 1, \ldots, m$ used to compute the non-linearity measures are taken on the linear theory confidence ellipsoid

$$\begin{align*}
(\theta - \hat{\theta})' X' X (\theta - \hat{\theta}) &= d^2 \\
(2.6)
\end{align*}$$

where $d^2 = ps^2 F_{p, \nu; \alpha}$. (In all of the examples of this section $S(\hat{\theta})/(n-p)$ is used for $s^2_{\nu}$, and $\alpha = .05$). Since Beale's non-linearity measures are unaffected by linear transformations of parameters, he finds it convenient to make a transformation

$$\theta = \hat{\theta} + Qt, \quad (2.7)$$

where $Q$ is $p \times p$, such that in $t$-space $(2.6)$ is given by

$$t' t = 1. \quad (2.8)$$

To estimate $N_\theta$ and $N_\phi$, it is necessary to derive a quadratic
Figure 2.7 The curves $\eta_3 (\theta_1 \to \infty, \theta_2 = .4, \xi)$ and $\eta_3 (\theta_1 = 1.4, \theta_2 = .4, \xi)$. 
approximation to \( \eta_u = f(\theta, \xi_u) \), \( u = 1, \ldots, n \), in the neighborhood of \( \theta = \hat{\theta} \), i.e., in the neighborhood of \( \mathbf{t} = 0 \). Denote \( f(\theta, \xi_u) \) by \( f_u \). If we make the expansion

\[
f_u = f_{u0} + \sum_{j=1}^{p} c_{uj} t_j + \sum_{k=1}^{p} c_{ujk} t_j t_k, \tag{2.9}
\]

where \( c_{ujk} = c_{ukj} \), then the formula for the theoretical measure of the non-linearity of the model is

\[
N_\theta = \frac{\sigma^2}{\theta (p+2) d^4} \sum_{u=1}^{n} \sum_{j=1}^{p} \sum_{k=1}^{p} \left( c_{ujj} c_{ukk} + 2c_{ujk}^2 \right). \tag{2.10}
\]

Also, if we define

\[
c_{ujk}^* = c_{ujk} - \sum_{v=1}^{n} p_{uv} c_{vjk}, \tag{2.11}
\]

where

\[
p_{uv} = \frac{1}{d^2} \sum_{i=1}^{p} c_{ui} c_{vi},
\]

then the theoretical measure of the intrinsic non-linearity is

\[
N_\phi = \frac{\sigma^2}{\phi (p+2) d^4} \sum_{u=1}^{n} \sum_{j=1}^{p} \sum_{k=1}^{p} \left( c_{ujj}^* c_{ukk}^* + 2c_{ujk}^2 \right). \tag{2.12}
\]

In the expression \( (2.9) \), \( f_{u0} \) is of course given by

\[
f_{u0} = f(\hat{\theta}, \xi_u), \quad u = 1, \ldots, p.
\]

From the linear transformation \( (2.7) \), the coefficients \( c_{uj} \) are given by

\[
c_{uj} = \sum_{i=1}^{p} \left( \frac{\partial f_u}{\partial \xi_1} \right)_{\xi_0} q_{ij}, \tag{2.13}
\]

where the \( \frac{\partial f_u}{\partial \theta_1} \) can be estimated by difference quotients. To obtain estimates of the \( c_{ujk} \), we need to compute \( f_u \) at \( p (p+1)/2 \) points in parameter space. Beale recommends using \( p(p+1) \) points, so
that the arrangement of points about \( \hat{\theta} \) can be made symmetric, and placed so that no cubic term in the expansion of \( f_u \) can have any effect on the estimates of the \( c_{ujk} \). The formulae for the \( c_{ujk} \) are given in Beale (1960 b), and also stated in Meeter (1964).

Besides comparing the non-linearity measures \( N_\theta \) and \( N_\phi \) with \( \hat{N}_\theta \) and \( \hat{N}_\phi \), there is one further comparison that we make. In computing the contours based on first and second order Taylor Series expansions of \( \eta_2 \) and \( \eta_3 \), we used the analytically available partial derivatives

\[
\frac{\partial f(\theta, \xi_u)}{\partial \theta_j}, \quad \frac{\partial^2 f(\theta, \xi_u)}{\partial \theta_i \partial \theta_k}, \quad j, k = 1, \ldots, p \quad u = 1, \ldots, n,
\]
evaluated at \( \hat{\theta} \). Thus, we can replace the difference quotients used to compute the \( c_{uj} \) (2.13) by the exact first derivatives. Also, by definition of the linear transformation from \( \theta \) to \( \xi \), (2.7), we have exact values \( c_{ujk} \) from

\[
c_{ujk} = \frac{\partial^2 f}{\partial \xi_i \partial \xi_k} = \sum_{i=1}^{p} \sum_{h=1}^{p} \frac{\partial^2 f(\theta, \xi_u)}{\partial \theta_i \partial \theta_h} \left[ \hat{\theta} \right]^{ij} \hat{\xi}^{hk}
\]

We will denote by \( \hat{N}_\theta \) and \( \hat{N}_\phi \) the non-linearity measures obtained by substituting the exact values of \( c_{uj} \) and \( c_{ujk} \) into the formulae (2.10) and (2.12).
In this case, \( p=2 \) so that for each example we have \( p(p+1)=6 \) points to use to calculate the measures of non-linearity. These will be denoted by \( \tilde{N} \), \( \hat{N} \), and \( \check{N} \), where

(i) the \( \tilde{N} \) measures are obtained from (2.10) and (2.12) using the analytic partial derivatives for the \( c \)'s,

(ii) the \( N \) measures are obtained from (2.10) and (2.12) using estimates of the \( c \)'s (see the Appendix), and

(iii) the \( \hat{N} \) measures are obtained from (1.5) and (1.8).

The six points are located on the ellipse (2.6), but because of the scaling convention adopted here, i.e., the parameter space is scaled in units of the linear theory standard derivations of the parameter estimates, in general none of the points will be at the endpoints of the principal axes of the ellipse in the unscaled \( \theta \)-space. These points are plotted in Figures 2.3 to 2.6 for the model \( \eta_2 \), with \( n=4 \), and \( \eta_3 \) with \( n=4, 6, 12 \).

Note that the ellipse for the model \( \eta_3 \) is so attenuated that only a few of the points can be plotted for the cases of four and six observations. Most of the other points are such that \( \theta_1 \) or \( \theta_2 \) is negative, which can lead to extremely high values of \( S(\theta) \) since the restriction \( 0 < \eta < 1 \) no longer holds. In these cases, we would expect that the assumptions underlying the derivations of Beale's \( N \) and \( \check{N} \) measures would break down. Table 2.1 demonstrates that this is

<table>
<thead>
<tr>
<th>Table 2.1</th>
<th>Non-linearity measures for the examples with four observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>MODEL</td>
<td>( \tilde{N}_\theta )</td>
</tr>
<tr>
<td>------</td>
<td>-----------------</td>
</tr>
<tr>
<td>( \eta_2 )</td>
<td>0.0208</td>
</tr>
<tr>
<td>( \eta_3 )</td>
<td>12.41</td>
</tr>
</tbody>
</table>

* -- greater than 1000.
so for the case \( n = 4 \). The two columns at the right of the table contain Beale’s suggested limits for assessing the significance of a particular value of the \( \hat{N} \) or \( \hat{\bar{N}} \) measures. (Recall that if \( \hat{N}_\theta > \frac{1}{F} \), where in our case \( F = F_{p, n-p; 0.05} \), the root mean square value of the discrepancy vector is greater than the intended vector, while if \( \hat{N}_\theta < \frac{0.01}{F} \), the root mean square value of the discrepancy vector is less than one-tenth of the length of the intended vector.)

Some explanation should be made of the extremely low values for the \( \hat{N} \) measures. If \( d \) is the sample space distance from \( \theta \) to any point on the ellipse (2.6), then, as was shown in the footnote to (1.16), aside from a factor \( p s^2 \), the numerator of \( \hat{N}_\theta \) is a sum of terms of the form \( \epsilon^2 d^2 \), each representing the squared length of a discrepancy vector, while the denominator is a sum of terms of the form \( (d^2 + (\epsilon d)^2 - 2\epsilon d^2 \cos \alpha)^2 \), the fourth power of the length of the actual vector. Now when the squared distance \( S(\theta) \) is several orders of magnitude larger than either \( S(\hat{\theta}) \) or \( d^2 \) (see Figure 2.8), the ratio \( \epsilon^2 d^2 / (d^2 + (\epsilon d)^2 - 2\epsilon d^2 \cos \alpha) \) is nearly one, and \( (\epsilon d)^2 / d^2 = \epsilon^2 \) is large. Since \( \epsilon \) appears to the second power in the numerator of \( \hat{N}_\theta \) and to the fourth power in the denominator, any one point representing a sufficiently radical departure from linearity will cause the \( \hat{N} \) measures to seriously underestimate the non-linearity. (Only one point is required since the factor \( d^2 \) is constant for each of the points used to compute \( \hat{N}_\theta \).)
Figure 2.8  The case when $S(\theta)$ is much larger than $S(\hat{\theta})$ and $d^2$. The labels are in terms of squared distances.

Referring again to Table 2.1, we note that for the model $\eta_3$, the $N$ measures are overwhelmingly large. Again, this is the result of the extreme curvature of the solution locus in the regions where $\theta_1$ or $\theta_2$ is negative, indicated by the disproportionately high values of $S(\theta)$. High values of $S(\theta)$ imply high values of $\eta = f(\theta, \xi_u)$ so that the $c_{ijk}$ in (2.9), which are estimated from linear combinations of the $f(\theta, \xi)$, [see Beale (1960 b) and Meeter (1964)] can become extremely large.

In Table 2.2, we summarize the results for the case of six observations.

<table>
<thead>
<tr>
<th>MODEL</th>
<th>$\eta_2$</th>
<th>$\eta_3$</th>
<th>$\tilde{N}_\theta$</th>
<th>$\tilde{N}_\phi$</th>
<th>$N_\theta$</th>
<th>$N_\phi$</th>
<th>$\hat{N}_\theta$</th>
<th>$\hat{N}_\phi$</th>
<th>$L/F$</th>
<th>.01/F</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\eta_2$</td>
<td>0.109</td>
<td>0.031</td>
<td>0.028</td>
<td>0.009</td>
<td>0.029</td>
<td>0.0010</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\eta_3$</td>
<td>7.355</td>
<td>0.173</td>
<td>76.51</td>
<td>8.442</td>
<td>0.0000</td>
<td>0.0000</td>
<td>1.144</td>
<td>0.00144</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The $N$ and $\hat{N}$ measures for the model $\eta_3$ exhibit essentially the same characteristics as for the four observation case, as we would expect from a comparison of Figures 2.3 and 2.5. The $N$ and $\hat{N}$ measures for the $\eta_2$ model, on the other hand, are near the value $.01/F$ representing "low" non-linearity, and agree quite well with each other.

The results for the case of 12 observations are contained in Table 2.3. The model $\eta_2$ is now essentially linear within the region corresponding to the linear theory 95% confidence ellipsoid. The $N$ measures for the model $\eta_3$ are still drastically underestimating the non-linearity, even though the points used in the computations are well within the region of positive values of $\theta_1$ and $\theta_2$. However, the argument
Table 2.3

Non-linearity measures for the examples with twelve observations

<table>
<thead>
<tr>
<th>MODEL</th>
<th>$\eta_2$</th>
<th>$\eta_3$</th>
<th>$\eta_4$</th>
<th>$N_0$</th>
<th>$N_\phi$</th>
<th>$N_\theta$</th>
<th>$N_\phi$</th>
<th>$\hat{N}_0$</th>
<th>$\hat{N}_\phi$</th>
<th>$1/F$</th>
<th>$.01/F$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\eta_2$</td>
<td>0.0037</td>
<td>0.0011</td>
<td>0.0009</td>
<td>0.0003</td>
<td>0.0007</td>
<td>0.0002</td>
<td>.244</td>
<td>.00244</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\eta_3$</td>
<td>1.421</td>
<td>0.063</td>
<td>6.129</td>
<td>0.115</td>
<td>0.0221</td>
<td>0.0001</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

cconcerning the behavior of the $\hat{N}$ measures still applies, i.e., we can expect the $\hat{N}$ measures to underestimate the non-linearity when $S(\theta)$ is much larger than $S(\hat{\theta})$ or $d^2$. For Table 2.3 and the model $\eta_3$, the relevant quantities are $S(\hat{\theta}) = .0067$, and $d^2 = \frac{p S(\hat{\theta})}{(n-p) F_p, n-p; .05} = .0055$. The actual sum of squares $S(\hat{\theta})$ for one of the six points was .0789. (The sum of squares predicted by linear theory for that point is of course $S(\hat{\theta}) + d^2 = .0122$.)

Finally, then, we conclude that the $\hat{N}$ measures are unreliable in the presence of high non-linearity, in the sense that they err in the direction of under-estimating the non-linearity. However, for models such as $\eta_2$, where the actual sum of squares $S(\theta)$ for the calculated points were never more than twice as large as the value predicted by linear theory, the $\hat{N}$ measures agreed well with the $N$ measures. Of course, the actual sums of squares should in any case be compared with the linear theory sum of squares, whether or not any non-linearity measures are calculated.

Both the $N$ measures and, with the above qualification, the $\hat{N}$
measures seem to reflect fairly well the overall picture of non-linearity, judging by the few examples we have encountered here. We do feel that the rough upper bound .01/F, representing the measure of non-linearity a model can have and still be considered essentially linear, is somewhat conservative, considering the performance of the model $\eta^2$.

Also, recall from section 1. that the quantities $N_\phi$ or $\hat N_\phi$ could be used to insure that the various tests and confidence regions had an associated probability greater than or equal to $1 - \alpha$. In view of the high variability among the various measures, this practice, though useful to increase the validity of the approximate statistical theory, is not very precise unless $N_\phi$ and $\hat N_\phi$ are less than, say .01/F, which would make the actual corrections very small. In any case, the exact probability level of tests and confidence regions is probably of less interest to an experimenter convinced of the validity and reasonableness of the Bayesian* approach, than to a person who is of the classical (that is, sampling theory) persuasion. As remarked by Barnard (1960), in the discussion to Beale's (1960a) paper, "...this measure will be appropriate only when the departures from linearity are reasonably small. When this is not the case there will be nothing for it but to explore the whole likelihood surface ...."

* It is of interest to note that Beale (1960a) described himself as "a Bayesian of the Jeffreys school."

The procedure for minimizing non-linearity in the neighborhood of $\hat{\theta}$ by transformations of parameters is basically the same as the procedure used in I. In this section, however, we will be minimizing the non-linearity at more than one point around $\hat{\theta}$ and the function to be minimized will be of a different form than $N(b)$ of I.

Suppose that the transformation $\varphi = \varphi(\theta)$ satisfies the conditions (1.6), and that the specific values $\varphi_w$, $w = 1, \ldots, m$ which minimize $Q_\theta$ as given by (1.2), have been computed using the normal equations (1.7). If the points $\theta_w$ used to compute $Q_\theta$ are kept fixed, then we minimize $\hat{N}_\theta$ by minimizing $Q_\theta$, the numerator of $\hat{N}_\theta$ apart from the constant factor $p_s$.* The "best" we can possibly do by a transformation of parameters is to reduce $\hat{N}_\theta$ to $\hat{N}_\varphi$, that is, reduce $Q_\theta$ to $Q_\varphi$, the value of $Q_\theta$ when $\varphi_w$ is substituted for $\theta_w$ in $Q_\theta$.

*There may be some question as to our reasons for minimizing the $\hat{N}$ measure rather than the $N$ measure, in view of the unsatisfactory performance of the former in the presence of high values of $S(\theta)$. We worked with the $\hat{N}$ measure rather than the $N$ measure, because we found it easier to express the relevant formulas for $\hat{N}$ in a way which allowed usage of the iterative least squares procedures described in I. A point still to be investigated is the conditions under which minimizing either of the measures would lead to essentially the same transformation.
Rather than minimize $Q_\theta$ with the transformation $\varphi = \phi(\theta)$, equivalently we can minimize $Q_\theta - Q_\phi$, which, as pointed out by Beale, gives a simpler formulation to the function to be minimized. Suppose we write $Q_\phi$ for $Q_\theta$ when $\varphi = \phi(\theta)$ replaces $\theta - \hat{\theta}$ in $Q_\theta$. [We recall that $\varphi = \phi(\theta)$ satisfies (1.6).] Letting $\eta_w = \eta(\theta_w)$, $\hat{\eta} = \eta(\hat{\theta})$, we can write, using matrix notation, (1.2), and the formula for $Q_\phi$ mentioned after (1.6), we have

$$Q_\varphi - Q_\phi = \sum_{w=1}^{m} \{ (\eta_w - \hat{\eta} - X_{\varphi_w})' (\eta_w - \hat{\eta} - X_{\phi_w}) $$

$$- (\eta_w - \hat{\eta} - X_{\phi_w})' (\eta_w - \hat{\eta} - X_{\phi_w}) \} $$

$$= \sum_{w=1}^{m} \{ (\eta_w - \hat{\eta})' (\eta_w - \hat{\eta}) - 2\varphi_w X' (\eta_w - \hat{\eta}) + \varphi_w X' X_{\varphi_w}$$

$$- (\eta_w - \hat{\eta})' (\eta_w - \hat{\eta}) + 2\phi_w X' (\eta_w - \hat{\eta}) - \phi_w X' X_{\phi_w} \} ,$$

and hence we obtain for $Q_\varphi - Q_\phi$, the expression

$$\sum_{w=1}^{m} \{ \varphi_w X' X_{\varphi_w} - 2(\varphi_w - \phi_w)' X' (\eta_w - \hat{\eta}) - \phi_w X' X_{\phi_w} \}. \quad (3.1)$$

But from the normal equations (1.7) for $\phi_w$, the second term within curly brackets in (3.1) can be written as

$$-2(\varphi_w - \phi_w)' X' X_{\varphi_w}$$

$$= -2\varphi_w' X' X_{\varphi_w} + 2\phi_w' X' X_{\phi_w}$$
so that

\[ Q_\phi - Q_{\bar{\phi}} = \sum_{w=1}^{m} \{ \phi_i' X'X \phi_i - 2\phi_i' X'X \phi_w + \phi_i' X'X \phi_w \} \]

\[ = \sum_{w=1}^{m} \{ (\phi_i - \phi_w)' X'X (\phi_i - \phi_w) \} \]

\[ = \sum_{i=1}^{p} \sum_{j=1}^{p} m_{ij} \{ \sum_{w=1}^{m} (\phi_{iw} - \phi_{1w}) (\phi_{jw} - \phi_{jw}) \}, \quad (3.2) \]

where \( m_{ij} \) is the \( i,j \)th element of the \( p \times p \) matrix \( X'X \).

Consider now the class of transformations

\[ \phi_i = (\theta_i + a_i)b_i, \quad a_i > \left| \theta_i \right|, \quad i = 1, \ldots, p, \quad (3.3) \]

where the \( \theta_i \) considered here are, for our purposes, in the vicinity of \( \hat{\theta} \). If we wish to minimize (3.2) within the class of transformations (3.3), in order to observe the condition (1.6), we must replace \( \phi_{kw}, k=1, \ldots, p \) in (3.2) by
\[ \psi_{kw}(a_k, b_k) = \frac{\varphi_{kw}(a_k, b_k)}{\frac{\partial \varphi_{kw}(a_k, b_k)}{\partial \theta_k}} \bigg|_{\theta_k} \]

which in our case is given by

\[
\frac{(b_k^* + a_k^*)^k - (\hat{\theta}_k + a_k^*)^k}{b_k^{k-1}(\hat{\theta}_k + a_k^*)}, \quad k = 1, \ldots, p. \tag{3.4}
\]

There are a number of ways to derive normal equations to approximately minimize \((3.1)\) as a function of \(a\) and \(b\), one of which is the following. Write

\[
\psi_{kw}(a_k, b_k) = \psi_{kw}(a_{ko}, b_{ko}) + (a_k - a_{ko}) \frac{\partial \psi_{kw}}{\partial a_k} \bigg|_{a_{ko}}
\]

\[
+ (b_k - b_{ko}) \frac{\partial \psi_{kw}}{\partial b_k} \bigg|_{b_{ko}}, \tag{3.5}
\]

where \(a_{ko}, b_{ko}, k = 1, \ldots, p\) are our initial guesses for the "best" transformation. Put \(\alpha = a - a_o\), \(\beta = b - b_o\), and

\[ r_{kw} = \psi_{kw}(a_{ko}, b_{ko}) - \phi_{kw}. \]

Then \((3.4)\) is approximately minimized by minimizing (as a function of \(\alpha\) and \(\beta\))

\[
Q(\alpha, \beta) = \sum_{i=1}^{p} \sum_{j=1}^{p} \sum_{m} \sum_{w=1}^{m} (\alpha_i c_{iw} + \beta_i d_{iw} + r_{iw})(\alpha_j c_{jw} + \beta_j d_{jw} + r_{jw}), \tag{3.6}
\]

where, suppressing the subscripts 'i' and 'w',

\[
c_{iw} = \frac{\partial \psi_{iw}}{\partial a_1} = \frac{1}{b(\hat{\theta} + a)^{b-1}} \left[ b(\theta + a)^{b-1} - (\hat{\theta} + a)^{b-1} - (\theta + a)^{b-1} \right], \tag{3.7}
\]

and
\[ d_{iW} = \frac{\partial \psi_{iW}}{\partial b_i} = \frac{1}{b(\hat{\theta} + a)^{b-1}} \left[ (\theta + a)^b \ln \left( \frac{\theta + a}{\hat{\theta} + a} \right) - \frac{1}{b} \{ (\theta + a)^b - (\hat{\theta} + a)^b \} \right] \]

Let \( C_w \) and \( D_w \) be \( p \times p \) diagonal matrices whose \( i \)th diagonal elements are \( c_{iw} \) and \( d_{iw} \), respectively, and let \( r_w \) be the \( p \times 1 \) vector whose \( i \)th element is \( r_{iw} \). Then \( Q(\alpha, \beta) \) can be written as

\[
Q(\alpha, \beta) = \sum_{w=1}^{m} \left[ r_w + C_w \alpha + D_w \beta \right]^T X^T X \left[ r_w + C_w \alpha + D_w \beta \right].
\]

Now it is well known that the minimum of a quadratic form such as

\[(y - Z \theta)' W (y - Z \theta),\]

as a function of \( \theta \), is given by

\[
\hat{\theta} = (Z' W Z)^{-1} Z' W y,
\]

where \( \theta \) is \( p \times 1 \), \( Z \) is \( n \times p \), \( y \) is \( n \times 1 \), and \( W \) is positive definite, and of order \( n \times n \). Then, in place of \( W \) in (3.8) we have

\[
\begin{bmatrix}
X' X & 0 & 0 & \cdots & 0 \\
0 & X' X & 0 & \cdots & 0 \\
\vdots & \vdots & \ddots & \ddots & \vdots \\
0 & 0 & \cdots & 0 & X' X
\end{bmatrix}
= \begin{bmatrix} \mathbb{I}_m \otimes X' X \end{bmatrix}_{pm \times pm}
\]

and where \( y \) corresponds to

\[
\begin{bmatrix}
\ell_l \\
\vdots \\
\ell_w \\
\vdots \\
\ell_m
\end{bmatrix}_{pm \times 1}
\]
and \( Z_2 \) is replaced by
\[
\begin{bmatrix}
C_1 & D_1 \\
\vdots & \\
C_m & D_m
\end{bmatrix}
\begin{bmatrix}
\alpha \\
\beta
\end{bmatrix}_{2p \times 1}
\]
Thus, applying (3.9),
\[
\begin{bmatrix}
\hat{\alpha} \\
\hat{\beta}
\end{bmatrix} = - \left( \begin{bmatrix}
C_1 & \cdots & C_m \\
D_1 & \cdots & D_m
\end{bmatrix}
\begin{bmatrix}
I_m \otimes X'X \\
C_m & D_m
\end{bmatrix} \right)^{-1}
\times
\begin{bmatrix}
C_1 & \cdots & C_m \\
D_1 & \cdots & D_m
\end{bmatrix}
\begin{bmatrix}
I_m \otimes X'X \\
C_m & D_m
\end{bmatrix}
\]
\tag{3.10}

The matrix to be inverted is then of the form
\[
\begin{bmatrix}
M_{cc} & M_{cd} \\
\cdots & \cdots \\
M_{dc} & M_{dd}
\end{bmatrix}
\]
\tag{3.11}

where
\[
M_{cd} = \sum_{w=1}^{m} C_w X'X D_w,
\]
\[
M_{dc} = M_{cd}' = \sum_{w=1}^{m} D_w X'X C_w,
\]
etc. That is, the \( i, j \)th element of, say, \( M_{cd} \) is given by
\[
m \sum_{jw=1}^{m} C_{iw} d_{jw}.
\]
The remainder of the right hand side of (3.10) is the vector
\[
\begin{bmatrix}
\sum_{w=1}^{m} C_w X'X r_w \\
\cdots \\
\sum_{w=1}^{m} D_w X'X r_w
\end{bmatrix}_{2p \times 1}
\]
in which, for example, the \( i \)th element \((i \leq p)\) in the vector can be expressed as

\[
\sum_{j=1}^{p} \sum_{w=1}^{m} c_{ijw} r_{iw}.
\]

Thus we can obtain \( a_i = \hat{a} + a_0 \), \( b_i = \hat{b} + b_0 \), substitute these values into \( Q_y - Q_\phi \), expand around \( a_i \) and \( b_i \) and continue the process until \( Q_y - Q_\phi \) stabilizes at some value which will usually be at least a local minimum of the empirical non-linearity measure at \( \hat{\theta} \) under the transformation (3.3).

The equations (3.10) for \( \hat{a} \) and \( \hat{b} \) are in a form which allows the method of Marquardt (discussed in 1) to be used. Unless there are strong prior considerations for a certain transformation, the most natural starting values for \( b \) would be \( b_{10} = 1, \ i = 1, \ldots, p \). However, we notice that the derivatives \( c_{iw} \) in (3.7) are zero at \( b=1 \) so that starting values for the \( b_i \) should be taken slightly different from 1 to avoid having the matrix (3.11) be singular. We can take \( a_{i0} = 0, \ i = 1, \ldots, p \), or large enough so that \( \theta_i + a_i \) is positive for all the points used to calculate \( Q_y - Q_\phi \), if we wish to consider negative values of \( b_i \).
As an example, we take the model

\[ \eta = \frac{\theta_1 \theta_2 \left( \xi_1^2 - \frac{\xi_2 \xi_3}{k} \right)}{(1 + \theta_2 \xi_1 + \theta_3 \xi_2)^2} \]  

(3.12)

a surface reaction controlling dual site rate equation for the reaction

\[ 2 \text{C}_2 \text{H}_5 \text{OH} \rightleftharpoons \text{C}_2 \text{H}_5 \text{OC}_2 \text{H}_5 + \text{H}_2 \text{O}, \]

where the reaction is postulated to occur between adjacently adsorbed alcohol molecules. The parameters are

\[ \theta_1 = \text{forward rate constant}, \]
\[ \theta_2 = \text{adsorption constant for alcohol, C}_2 \text{H}_5 \text{OH}, \]
and \[ \theta_3 = \text{adsorption constant for water, H}_2 \text{O}. \]

The independent variables are

\[ \xi_1 = \text{partial pressure of alcohol in the mixture}, \]
\[ \xi_2 = \text{partial pressure of water}, \]
and \[ \xi_3 = \text{partial pressure of ether, C}_2 \text{H}_5 \text{OC}_2 \text{H}_5. \]

The data are 18 observations taken from Kabel and Johanson (1962) where the overall equilibrium constant \( k \) in (3.12) was 25.2.

The estimates of the parameters which we obtained by the non-linear least squares procedure of I are

\[
\begin{array}{c|c|c|c}
\hat{\theta}_1 & \hat{\theta}_2 & \hat{\theta}_3 & S(\theta) \\
2.243 \times 10^{-4} & 3.402 & 6.146 & 7.999 \times 10^{-10} \\
\end{array}
\]  

(3.13)
From the equation

\[ S(\theta) = \frac{P}{n-p} S(\hat{\theta}) \mathbf{F}_{p,n-p; \alpha}, \quad (6.14) \]

with \( \alpha = .05 \), the value \( S = 1.198 \times 10^{-9} \) was obtained. If the model were truly linear, all of the points \( \theta \) on the ellipsoid

\[ S = S(\hat{\theta}) + (\theta - \hat{\theta})' \mathbf{X}' \mathbf{X} (\theta - \hat{\theta}), \]

where \( \mathbf{X} = \{ \frac{\partial f(\theta, \xi_u)}{\partial \theta_i} \} \), \( u = 1, \ldots, n, i = 1, \ldots, p \), would have sums of squares equal to \( S \). The actual sums of squares for the \( p(p+1) \) points on the ellipsoid \((3.15)\) used to calculate the non-linearity measures were either approximately equal to \( S \) or considerably larger, relative to \( S \) and \( S - S(\hat{\theta}) \), which would lead us to suspect that the \( \hat{N} \) measures would underestimate the non-linearity. The \( N \) and \( \hat{N} \) measures, calculated according to Sections 1 and 2, were

<table>
<thead>
<tr>
<th>( N_\theta )</th>
<th>( N_\phi )</th>
<th>( \hat{N}_\theta )</th>
<th>( \hat{N}_\phi )</th>
<th>( 1/F )</th>
<th>( .01/F )</th>
</tr>
</thead>
<tbody>
<tr>
<td>.3254</td>
<td>.0007</td>
<td>.0652</td>
<td>.0001</td>
<td>.4</td>
<td>.004</td>
</tr>
</tbody>
</table>

\((3.16)\)

This would indicate, according to Beale's criteria, that while the non-linearity was large enough to make linear theory results useful approximations only, there existed some transformation of parameters \( \phi = \phi(\theta) \) such that the model was essentially a linear function of these transformed parameters over the region \((3.15)\).

By applying the technique of this section, after four iterations, the transformation (taking \( \phi_i = (\theta_i + a_i) b_i, i = 1, \ldots, p \))
<table>
<thead>
<tr>
<th>$a_1$</th>
<th>$b_1$</th>
<th>$a_2$</th>
<th>$b_2$</th>
<th>$a_3$</th>
<th>$b_3$</th>
<th>$\hat{N}_\varphi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-1.6 \times 10^{-4}$</td>
<td>.74</td>
<td>-.32</td>
<td>-.38</td>
<td>-.35</td>
<td>-.11</td>
<td>0.0013</td>
</tr>
</tbody>
</table>

(3.17)

was determined. The value of $\hat{N}_\varphi$, even allowing for the fact that the $\hat{N}$ measures are lower than the $N$ measures by a factor of about seven, is reasonably close to the value .01/F. However, judging by the values of $\hat{N}_\varphi$ that were printed out during the convergence to (3.17), the estimates of the transformation were not particularly precise. For example, on the iteration previous to the one that produced (3.17) the results were:

<table>
<thead>
<tr>
<th>$a_1$</th>
<th>$b_1$</th>
<th>$a_2$</th>
<th>$b_2$</th>
<th>$a_3$</th>
<th>$b_3$</th>
<th>$\hat{N}_\varphi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-1.6 \times 10^{-4}$</td>
<td>.62</td>
<td>-.29</td>
<td>-.11</td>
<td>-.13</td>
<td>.24</td>
<td>0.0027</td>
</tr>
</tbody>
</table>

(3.18)

Because of these results, we decided to determine the effect of the transformation $a_1 = 0$, $i=1,2,3$, and $b_1 = 1$, $b_2 = -5$, $b_3 = 0$, or $\varphi_1 = 0_1$, $\varphi_2 = (\theta_2)^{-\frac{1}{2}}$, $\varphi_3 = \log \theta_3$. The choice of "no transformation" for $\theta_1$ was suggested by the relatively "weak" transformation .74 to which the procedure converged, and the fact that $\theta_1$ appears linearly in the model (3.12).

Figure 3.1 illustrates the contour $S(\theta) = S_2$ in $\theta_2$ and $\theta_3$ when $\theta_1 = \hat{\theta}_1$. The actual contour in the untransformed parameters does not
Figure 3.1 Contour of constant sum of squares in $\theta_2$ and $\theta_3$, when $\theta_1 = 2.24 \times 10^{-4}$
Figure 3.2 The contour of Figure 4.3.1, when plotted uniformly in scales of $(\theta_2)^{\frac{1}{2}}$, $\log \theta_3$.

$\theta_2 = 3.40$, $\theta_3 = 6.15$
appear to indicate serious non-linearity. However, it is clear that no ellipsoid centered at \( \hat{\theta} \) will be able to approximate contours of constant \( S(\theta) \) very well. Figure 3.2 illustrates the same contour when plotted linearly in \( \varphi_2 = (\theta_2)^{-\frac{1}{2}} \), \( \varphi_3 = \log \theta_3 \). The contour is now satisfactorily elliptical. Since the range of \( \theta_2 \) is fairly small, it is quite likely that other transformations of \( \theta_2 \), such as \( \varphi_2 = \log \theta_2 \), would give almost the same results.

Assuming that the contours for other relevant values of \( \theta_1 \) reveal essentially the same characteristics, (this was in fact found to be the case) the assumption that the model is locally linear in \( \varphi \) is now a very good approximation. That is, if \( \Delta \) is a \( p \times p \) diagonal matrix whose \( i^{th} \) diagonal element is \( \frac{\partial \theta_i}{\partial \varphi_1} \bigg|_{\varphi(\hat{\theta})} \), it is essentially true that

\[
\eta = \eta(\hat{\theta}) + X \Delta (\varphi - \varphi(\hat{\theta}))
\]

within a region in \( \varphi \)-space bounded by

\[
S = (\varphi - \varphi(\hat{\theta}))' \Delta X' X \Delta (\varphi - \varphi(\hat{\theta}))
\]

with all of the resulting simplification of interpretation that this implies.
References


