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ANALYSIS OF MIXTURE EXPERIMENTS IN PRESENCE OF BLOCK EFFECTS

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SUMMARY. In the presence of block effects the estimates of the regression parameters are obtained by imposing an appropriate linear restriction on the block parameters. Orthogonal blocking is also possible under the restriction $\Sigma\, n_w \gamma_w = 0$ and suitably chosen blocking conditions.

1. Introduction

Scheffe (1958, 1963) and Murty and Das (1968) evolved a number of designs for mixture experiments. Box and Hunter (1957) stated that blocking is a desirable property of any response design, as it is an important tool for obtaining increased precision. Murty (1966) reached the conclusion which was, however, based on emperical reasoning that actual blocking is not possible in the symmetric simplex designs without replicating the design itself. Nigam (1970) derived conditions for estimating the parameters of a quadratic model in presence of block effects. Saxena and Nigam (1975) constructed designs satisfying these conditions. Later Nigam (1976) found that under these blocking conditions the design matrix is singular, and suggested corrections to the blocking conditions. But the new conditions of Nigam (1976) as well do not remove the singularity of the design matrix. The design matrix still remains singular and estimates of parameters under these conditions are not unique.

The estimation of the parameters by adjusting for block effects is possible, only with suitable assumption on the block parameters. It has been shown in Section 6 that under blocking conditions of Nigam (1970), orthogonal blocking can be achieved, provided the restriction of the form

$$\sum_w n_w \gamma_w = 0$$

is imposed on the block parameters.

2. Quadratic model in presence of block effects

Let n design points be arranged in t blocks such that the w-th block contains n_w ($w = 1, \ldots, t$) points and $\sum_w n_w = n$. Let γ_w denote the effect of the w-th block. A quadratic model in k variables in presence of block effects can be written as

$$y_u = \sum_{i=1}^{k} \beta_i x_{iu} + \sum_{i<j} \beta_{ij} x_{iu} x_{ju} + \sum_w \gamma_w z_{wu} + e_u, \qquad \ldots \quad (1)$$

where

(i) $E(e_u) = 0$,

(ii) $E(e_u e_{u'}) = 0, \quad$ if $u \neq u'$

$\qquad = \sigma^2, \quad$ if $u = u'$

(iii) $Z_{wu} = 1$, if u-th point lies in the w-th block

$\qquad = 0$, otherwise.

3. Normal equations under the symmetry conditions and blocking conditions

We assume that the design satisfies the symmetry conditions of Murty and Das (1968), viz.,

$$\left.\begin{aligned}
&\sum_u x_{iu}^2 = A, \\
&\sum_u x_{iu} x_{ju} = B, \\
&\sum_u x_{iu}^2 x_{ju} = C, \\
&\sum_u x_{iu} x_{ju} x_{ku} = D, \\
&\sum_u x_{iu}^2 x_{ju}^2 = E, \\
&\sum_u x_{iu}^2 x_{ju} x_{ku} = F, \\
\text{and} \quad &\sum_u x_{iu} x_{ju} x_{ku} x_{lu} = G,
\end{aligned}\right\} \qquad \ldots \quad (2)$$

where A, B, C, D, E, F and G are constants for all $i \neq j \neq k \neq l$, and $u = 1, 2, \ldots, n$.

We further assume the blocking conditions of Nigam (1976) viz.,

$$\sum_{u=1}^{n_{w}} x_{iu} = K_{1w} \text{ (constant)}$$

and (3)

$$\sum_{u=1}^{n_{w}} x_{iu}x_{ju} = K_{2w} \text{ (constant)}.$$

for all $w = 1, 2, \ldots, t$ and

$$i \neq j, \; i, j = 1, 2, \ldots, k.$$

The normal equations following from model (1) for a design satisfying the above conditions are as below :

$$(A-B)\hat{\beta}_{\lambda}+B\sum_{i}\hat{\beta}_{i}+(C-D)\sum_{j \neq \lambda}\hat{\beta}_{\lambda j}+D\sum_{i<j}\hat{\beta}_{ij}+\sum_{w}K_{1w}\hat{\gamma}_{w}$$

$$= \sum_{w=1}^{t}\left(\sum_{u=1}^{n_{w}} x_{\lambda u}y_{u}\right). \quad \ldots \quad (4)$$

for $\lambda = 1, 2, \ldots, k$.

$$(C-D)(\hat{\beta}_{\lambda}+\hat{\beta}_{\mu})+D\sum\hat{\beta}_{i}+(E-2F+G)\hat{\beta}_{\lambda\mu}+(F-G)\left(\sum_{j \neq \lambda}\hat{\beta}_{\lambda j}+\sum_{j \neq \mu}\hat{\beta}_{\mu j}\right)$$

$$+G\sum_{ij}\hat{\beta}_{ij}+\sum_{w}K_{2w}\hat{\gamma}_{w} = \sum_{w=1}^{t}\left(\sum_{u=1}^{n_{w}} x_{\lambda u}x_{\mu u}y_{u}\right) \quad \ldots \quad (5)$$

for $\gamma, \mu = 1, 2, \ldots, k$; and $\lambda \neq \mu$, and

$$K_{1w}\sum_{i}\hat{\beta}_{i}+K_{2w}\sum_{i<j}\hat{\beta}_{ij}+n_{w}\hat{\gamma}_{w} = \sum_{u=1}^{n_{w}} y_{u} = B_{w}\text{(say)}, \; w = 1, 2, \ldots, t.$$

$$\ldots \quad (6)$$

It can be easily shown that under the modified blocking conditions of Nigam as given in (3), the design matrix is still singular.

4. Estimation of parameters

In order to obtain a unique solution of the normal equations some restriction on the block parameters is essential. The implication of this assumption is that the block effects, γ_{w}'s and the regression parameters of the type β_{i}'s can not be estimated uniquely, but their contrasts can be estimated. The set of equations which provide the optimum combination of the mixture variables involves only the contrasts of β's. Thus the estimation of optimum combination of components does not suffer from the lack of uniqueness of the estimates of regression parameters. It is, however, of interest to find that the estimate of y based on the prediction equation

$$\hat{y} = \sum \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j$$

depends upon the assumption on the block parameters, and should be taken as an estimate of y for the circumstances under which the assumption is appropriate. For example, if it is desired to have an estimate under condition of experiment represented by average block effect, the reasonable assumption is

$$\sum_{w} n_w \gamma_w = 0$$

and, if the estimate is required for experimental conditions of the w-th block, the assumption should be

$$\gamma_w = 0.$$

In this section we shall obtain estimates of regression parameters by adjusting for block effects under the symmetry conditions (2), blocking conditions (3) and the assumption

$$\sum_{w} n_w \gamma_w = 0. \qquad \dots \quad (7)$$

If the estimates of the regression parameters are required under some other assumptions, say

$$\sum_{w} a_w \gamma_w = 0, \qquad \dots \quad (8)$$

suitable adjustment to the estimates obtained in this section, may be applied. Such adjustments are discussed in Section 5. Using relations (6) and (7), equations (4) and (5) can be written as

$$(A-B)\beta_\lambda + B \sum_i \beta_i + (C-D) \sum_{j \neq \lambda} \hat{\beta}_{\lambda j} + D \sum_{i<j} \beta_{ij}$$

$$= \sum_{w=1}^{t} \left\{ \sum_{u=1}^{n_w} x_{\lambda u} y_u \right\} = \sum_{u=1}^{n} x_{\lambda u} y_u \text{ for } \lambda = 1, 2, \dots, k. \qquad \dots \quad (9)$$

and

$$(C^*-D^*)(\hat{\beta}_\lambda+\hat{\beta}_\mu)+D^* \sum_i \hat{\beta}_i+(B^*-2F^*+G^*)\hat{\beta}_\lambda$$

$$+(F^*-G^*)\left(\sum_{j\neq\lambda} \hat{\beta}_{\lambda j}+\sum_{j\neq\lambda} \hat{\beta}_{\mu j}\right)+G^* \sum_{i<j} \hat{\beta}_{ij}$$

$$= \sum_{w=1}^{t}\left(\sum_{u=1}^{n_w} x_{\lambda u}x_{\mu u}y_u\right)^*$$

$$= \left(\sum_{u=1}^{n} x_{\lambda u}x_{\mu u}y_u\right)^* \qquad \ldots \quad (10)$$

where

$$C^* = C-\sum_w \left(\frac{K_{1w}K_{2w}}{n_w}\right) = C-\frac{B}{k}$$

$$D^* = D-\sum_w \left(\frac{K_{1w}K_{2w}}{n_w}\right) = D-\frac{B}{k}$$

$$E^* = E-\sum_w \left(\frac{K_{2w}^2}{n_w}\right),$$

$$F^* = F-\sum_w \left(\frac{K_{2w}^2}{n_w}\right),$$

$$G^* = G-\sum_w \left(\frac{K_{2w}^2}{n_w}\right),$$

and

$$\left(\sum_u x_{\lambda u}x_{\mu u}y_u\right)^* = \sum_{u=1}^{n} x_{\lambda u}x_{\mu u}y_u - \sum_w \frac{K_{2w}B_w}{n_w},$$

where

$$\sum_{u=1}^{n_w} y_u = B_w.$$

The normal equations (9) and (10) are of the same form as those of Murty and Das (1968) except for some adjustment in the coefficients in the equations obtained by differentiating with respect to β_{ij}. The estimates of the parameters can be obtained by following the procedure of Murty and Das. The variances and covariances of the estimates of the regression parameters can be obtained by collecting the coefficients of appropriate terms in the solution of the estimates. It should, however, be noticed that $\frac{B}{n}$ will be the coefficient of Σy in $\sum_{w} \left(\frac{K_{2w}B_w}{n_w}\right)$.

5. Estimates and variances under the general assumptions

For obtaining the estimates of the regression parameters under the assumption (8), the estimates of the type β_i obtained in the previous section will be subject to an adjustment and the estimates of the type β_{ij} will remain the same.

Let $\hat{\beta}_i'$ $(i = 1, 2, \ldots, k)$ be the estimates under the assumption (8). Then we can easily obtain that

$$\hat{\beta}_i' = \hat{\beta}_i + \frac{\Sigma\, a_{iw}\hat{\gamma}_{iw}}{\Sigma\, a_{iw}}, \quad i = 1, 2, \ldots, k \qquad \ldots \quad (11)$$

where $\hat{\gamma}_{iw}$ is the estimate of γ_{iw} under (7) and is obtained from (6). Further

$$\hat{\gamma}_w' = \hat{\gamma}_{iw} - \frac{\Sigma\, a_w\hat{\gamma}_w}{\Sigma\, a_w}, \quad w = 1, 2, \ldots, l \qquad \ldots \quad (12)$$

where $\hat{\gamma}_w'$ is the estimate of γ_w under (8).

From (11) it is also clear that the estimates of the elementary contrast under the two assumptions are the same and hence the estimate is unique.

From (11) and (12) it is also obvious that the sum of squares for error and that for regression are the same for both the assumptions. This establishes the uniqueness of the regression sum of squares and error sum of squares.

The variances and covariances of the regression estimates under (8) can also be obtained by applying some adjustments to the variances and covariances of the regression estimates under (7).

As β_{ij} are uniquely estimated, their variances and covariances are also unique. Variances and covariances of other estimates can easily be obtained as below :

$$V(\hat{\beta}_i') = V(\hat{\beta}_i) + \Delta_1, \qquad \ldots \quad (13)$$

$$\text{cov}(\hat{\beta}_i', \hat{\beta}_j') = \text{cov}(\hat{\beta}_i, \hat{\beta}_j) + \Delta_1, \qquad \ldots \quad (14)$$

and

$$\text{cov}(\beta_i', \beta_{jl}') = \text{cov}(\beta_i, \beta_{jl})+\Delta_2 \quad \text{for } j \text{ or } l = i \text{ or } j \neq l \neq i \qquad \dots \quad (15)$$

where

$$\Delta_1 = \left[\left\{ \frac{1}{n} - \frac{\Sigma(a_w^2/n_w)}{(\Sigma a_w)^2} \right\} + \frac{k}{p} - \left\{ \frac{B^2}{n} - \frac{\Sigma\left(\frac{K_{2w}a_w}{n_w}\right)^2}{(\Sigma a_w)^2} \right\} \right] \sigma^2 \qquad \dots \quad (16)$$

$$\Delta_2 = \frac{2}{p(k-1)} \left[\frac{B}{n} - \frac{\Sigma\left(\frac{a_w K_{2w}}{n_w}\right)}{\Sigma a_w} \right] \sigma^2 \qquad \dots \quad (17)$$

and

$$p = E^* + 2(k-2)F^* + \frac{(k-2)(k-3)}{2} G^*.$$

6. Orthogonal blocking

Under the symmetry conditions (2) and the following blocking conditions

$$\frac{K_{1w}}{n_w} = K_1 \text{ (constant), for all } w = 1, 2, \dots, t$$

and

$$\frac{K_{2w}}{n_w} = K_2 \text{ (constant), for all } w = 1, 2, \dots, t \qquad \dots \quad (18)$$

and assumption (7), the normal equations become free from block effects and the regression parameters can be estimated by ignoring the block effects. In this sense, it may be regarded as an orthogonal blocking. The designs satisfying the above conditions have been constructed by Nigam (1970) and Saxena (1974).

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