D-optimal minimum support mixture designs in blocks

Goos, Peter and Donev, A.N.

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ORIGINAL ARTICLE

Peter Goos · Alexander N. Donev

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Abstract An easy method to construct efficient blocked mixture experiments in the presence of fixed and/or random blocks is presented. The method can be used when qualitative variables are involved in a mixture experiment as well. The resulting designs are $\mathcal{D}$-optimal in the class of minimum support designs. It is illustrated that the minimum support designs are more efficient than orthogonally blocked mixture experiments presented in the literature and only slightly less efficient than $\mathcal{D}$-optimal designs.

Keywords Fixed and random blocks · Minimum support design · Mixture experiment · Orthogonal blocking · Qualitative variables

1 Introduction

Most of the published work on blocked mixture experiments is on orthogonally blocked experiments which allow the mixture component effects to be estimated independently from the block effects. The conditions for the orthogonal blocking of mixture experiments were derived by Nigam (1976) and John (1984). They can be seen as special cases of the conditions established in Goos and Donev (2006a) which extend those proposed by Khuri (1992). Several examples of orthogonally blocked mixture designs are given in Cornell (2002). Draper et al. (1993) found orthogonally blocked mixture designs for experiments with four components and

P. Goos (✉)
Universiteit Antwerpen, Faculty of Applied Economics,
Prinsstraat 13, 2000 Antwerpen, Belgium
E-mail: peter.goos@ua.ac.be

A. N. Donev
AstraZeneca, Alderley Park,
Macclesfield, SK10 4TG, London, UK
E-mail: Alex.Donev@astrazeneca.com

two blocks, while Prescott et al. (1993; 1997) considered designs with five components. General methods for constructing orthogonally blocked mixture experiments with more than two components are presented by Lewis et al. (1994). Prescott and Draper (1998) derived orthogonally blocked designs for experiments with constrained components. Prescott (2000) showed how orthogonally blocked response surface designs can be projected onto a constrained design region in order to obtain an orthogonally blocked mixture design. Mixture designs that are not orthogonally blocked were derived by Donev (1989), who presented a simple method to construct efficient blocked mixture designs.

In all of these references, only the case of a single blocking variable that is treated as fixed was investigated. In some experimental situations, there is, however, more than one blocking variable. Typical examples of blocking variables are the vendor supplying the raw material, the shift or personnel running the experiments, the laboratory performing the experiments, or the day on which the runs are carried out. Clearly, some of these blocking variables should be treated as random. Other complications in many practical experiments are that the block sizes are small, so that designing orthogonally blocked experiments is often impossible. The aim of this paper is to propose a simple two-stage approach to design efficient blocked mixture experiments in such situations. The proposed designs are $\mathcal{D}$-optimal in the class of minimum support designs. For their construction, the simple design construction method presented by Donev (1989) is extended to the case of a blocking variable that is treated as random and to the case of more than one blocking variable.

The next two sections of this article are devoted to minimum support designs and blocked experiments. Next, the two-stage approach to construct $\mathcal{D}$-optimal minimum support designs in blocks is outlined. Finally, these designs are compared to orthogonally blocked experiments and to $\mathcal{D}$-optimal designs.

2 Minimum support designs

A minimum support design is a design which has observations at as many distinct combinations of the experimental variables as there are parameters in the statistical model that has to be estimated. Consider the following linear model:

$$y_i = \mathbf{f}'(\mathbf{x}_i)\boldsymbol{\beta} + \varepsilon_i, \quad i = 1, 2, \ldots, n, \tag{1}$$

where y_i is the response observed in the ith run of the experiment, $\mathbf{f}(\mathbf{x}_i)$ is the p-dimensional vector containing the model expansion of the settings of the experimental variables $\mathbf{x}$ at the ith observation, $\boldsymbol{\beta}$ is the p-dimensional vector containing the experimental variables' effects, ε_i represents the random error, and n is the number of observations. In matrix notation, this model is given by

$$\mathbf{y} = \mathbf{X}\boldsymbol{\beta} + \boldsymbol{\varepsilon}, \tag{2}$$

where $\mathbf{y}$ is the vector containing the n responses observed in the experiment, $\mathbf{X} = [\mathbf{f}(\mathbf{x}_1)\ \mathbf{f}(\mathbf{x}_2)\ \cdots\ \mathbf{f}(\mathbf{x}_n)]'$ is the $n \times p$ model matrix, and $\boldsymbol{\varepsilon}$ is the vector of random errors. We assume that $\boldsymbol{\varepsilon} \sim N(\mathbf{0}_n, \sigma_\varepsilon^2\mathbf{I}_n)$.

In this model, p parameters have to be estimated. A minimum support design for this model has therefore observations at only p distinct design points or support points $\mathbf{x}_i$. The model matrix $\mathbf{X}$ then only possesses p different rows. If we denote by n_i the number of times each of the support points $\mathbf{x}_i$ is used, then $\sum_{i=1}^{p} n_i = n$ and

$$\mathbf{X}'\mathbf{X} = \mathbf{X}_m'\mathbf{W}\mathbf{X}_m, \tag{3}$$

where $\mathbf{X}_m$ is a $p \times p$ matrix the rows of which are the model expansions of the p distinct support points and $\mathbf{W} = \text{diag}[\ n_1\ n_2\ \cdots\ n_p]$. Using this result, we find that

$$\mathbf{X}_m(\mathbf{X}'\mathbf{X})^{-1}\mathbf{X}_m' = \mathbf{X}_m(\mathbf{X}_m'\mathbf{W}\mathbf{X}_m)^{-1}\mathbf{X}_m' = \mathbf{X}_m\mathbf{X}_m^{-1}\mathbf{W}^{-1}(\mathbf{X}_m')^{-1}\mathbf{X}_m' = \mathbf{W}^{-1}, \tag{4}$$

for a nonsingular design. The elements of this $p \times p$ matrix are given by $\mathbf{f}'(\mathbf{x}_i)$ $(\mathbf{X}'\mathbf{X})^{-1}\mathbf{f}(\mathbf{x}_j)$ $(i, j = 1, 2, \ldots, n)$. Up to the proportionality constant σ^2, the diagonal elements are therefore nothing but the prediction variances in the support points of the design, and its off-diagonal elements are the covariances between the predictions for the design points. Equation (4) thus shows that the prediction variance for the ith design point is proportional to n_i^{-1} and that the covariance between the predictions for two distinct design points is zero. Remarkably, this result does not depend on the choice of the design points. It implies that, for any subset S of s support points,

$$\mathbf{1}_s'\mathbf{X}_s(\mathbf{X}'\mathbf{X})^{-1}\mathbf{X}_s'\mathbf{1}_s = \sum_{i \in S} n_i^{-1}, \tag{5}$$

and that for any two subsets S and T of s and t support points, respectively,

$$\mathbf{1}_s'\mathbf{X}_s(\mathbf{X}'\mathbf{X})^{-1}\mathbf{X}_t'\mathbf{1}_t = \sum_{i \in S \cap T} n_i^{-1}. \tag{6}$$

These results will be used extensively in the proofs of the theorems underlying the two-stage design construction approach presented in section 4.

3 Blocked experiments

In this section, we describe the statistical model corresponding to a blocked experiment and the criteria we utilize to evaluate the designs presented.

3.1 The statistical model

In a general setting with both fixed and random blocking variables, the statistical model corresponding to a blocked mixture experiment can be written as

$$\begin{aligned} \mathbf{y} &= \mathbf{X}\boldsymbol{\beta} + \mathbf{C}\boldsymbol{\gamma} + \mathbf{Z}\boldsymbol{\delta} + \boldsymbol{\varepsilon}, \\ &= \mathbf{F}\boldsymbol{\xi} + \mathbf{Z}\boldsymbol{\delta} + \boldsymbol{\varepsilon}, \end{aligned} \tag{7}$$

where $\mathbf{X}$ is the $n \times p$ extended design matrix corresponding to the components of the mixture, $\mathbf{C}$ is the design matrix corresponding to the indicator variables for the fixed blocks, $\mathbf{Z}$ is the design matrix corresponding to the indicator variables for the random blocks, and $\mathbf{F} = [\ \mathbf{X}\ \mathbf{C}\ ]$. The vectors $\boldsymbol{\beta}$, $\boldsymbol{\gamma}$, $\boldsymbol{\delta}$ and $\boldsymbol{\varepsilon}$ represent the mixture variable coefficients, the fixed block effects, the random block effects and the random errors, respectively. Finally, $\boldsymbol{\xi}' = [\ \boldsymbol{\beta}'\ \boldsymbol{\gamma}'\ ]$. We denote the number of fixed and random blocking variables by F and R, respectively, and the number of levels these variables possess by $b_{F,i}$ $(i = 1, 2, \ldots, F)$ and $b_{R,i}$ $(i = 1, 2, \ldots, R)$. Furthermore, we assume that $\boldsymbol{\varepsilon} \sim N(\mathbf{0}_n, \sigma_\varepsilon^2 \mathbf{I}_n)$, that $\boldsymbol{\delta} \sim N(\mathbf{0}_{b_R}, \mathbf{G})$, where $b_R = \sum_{i=1}^{R} b_{R,i}$ and

$$\mathrm{var}(\boldsymbol{\delta}) = \mathbf{G} = \mathrm{diag}\big(\sigma_1^2 \mathbf{I}_{b_{R,1}}, \sigma_2^2 \mathbf{I}_{b_{R,2}}, \ldots, \sigma_R^2 \mathbf{I}_{b_{R,R}}\big), \tag{8}$$

and that $\mathrm{cov}(\boldsymbol{\varepsilon}, \boldsymbol{\delta}) = \mathbf{0}_{n \times b_R}$. As a result,

$$\mathrm{var}(\mathbf{y}) = \mathbf{V} = \mathbf{ZGZ}' + \sigma_\varepsilon^2 \mathbf{I}_n. \tag{9}$$

For model (7) to be estimable, the matrix $\mathbf{F}$ has to be of full rank. This can be achieved by using only $b_{F,i} - 1$ indicator variables for the ith fixed blocking variable $(i = 1, 2, \ldots, F)$. When the random error terms as well as the random block effects are normally distributed, the maximum likelihood estimator of $\boldsymbol{\xi}$ in (7) is the generalized least squares estimator

$$\hat{\boldsymbol{\xi}} = \left[\hat{\boldsymbol{\beta}}'\ \hat{\boldsymbol{\gamma}}'\right]' = \left(\mathbf{F}'\mathbf{V}^{-1}\mathbf{F}\right)^{-1} \mathbf{F}'\mathbf{V}^{-1}\mathbf{y}, \tag{10}$$

the variance-covariance matrix of which is given by

$$\mathrm{var}(\hat{\boldsymbol{\xi}}) = \left(\mathbf{F}'\mathbf{V}^{-1}\mathbf{F}\right)^{-1}. \tag{11}$$

3.2 Design criterion

In the design of blocked experiments, the emphasis is on the computation of designs that allow an efficient estimation of the effects $\boldsymbol{\beta}$ of the experimental variables, rather than on the block effects $\boldsymbol{\gamma}$ and $\boldsymbol{\delta}$. In order to find such designs, we use the $\mathcal{D}$-optimality criterion which seeks designs that minimize the generalized variance of the parameter estimator $\hat{\boldsymbol{\beta}}$. This is done by minimizing the determinant of the part of (11) corresponding to $\boldsymbol{\beta}$, or, alternatively, by maximizing $|\mathbf{X}'\mathbf{V}^{-1}\mathbf{X} - \mathbf{X}'\mathbf{V}^{-1}\mathbf{C}(\mathbf{C}'\mathbf{V}^{-1}\mathbf{C})^{-1}\mathbf{C}'\mathbf{V}^{-1}\mathbf{X}|$. If the blocking structure is dictated by the experimental situation and cannot be determined by the experimenter, this is equivalent to maximizing the determinant of the information matrix $\mathbf{F}'\mathbf{V}^{-1}\mathbf{F}$ on the fixed effects $\boldsymbol{\xi}$. When R random blocking variables are involved in the experiment, the $\mathcal{D}$-optimal design depends on the ratios $\eta_1 = \sigma_1^2/\sigma_\varepsilon^2$, $\eta_2 = \sigma_2^2/\sigma_\varepsilon^2, \ldots, \eta_R = \sigma_R^2/\sigma_\varepsilon^2$ through $\mathbf{V}$. In order to compare two designs with model matrices $\mathbf{F}_1$ and $\mathbf{F}_2$ in terms of $\mathcal{D}$-efficiency, we use the relative $\mathcal{D}$-efficiencies $\{|\mathbf{F}_1'\mathbf{V}^{-1}\mathbf{F}_1|/|\mathbf{F}_2'\mathbf{V}^{-1}\mathbf{F}_2|\}^{1/p}$.

4 Design construction method

In this section we describe the two-stage approach to generate $\mathcal{D}$-optimal blocked minimum support designs. Theoretical justification for the approach is provided as well. The present results extend the one obtained by Donev (1989) who used (5) and (6) to show that, when there is one fixed blocking variable acting at b levels, $|\mathbf{F}'\mathbf{F}| = |\mathbf{X}'\mathbf{X}| \times |\mathbf{R}|$, where

$$\mathbf{R}=\begin{bmatrix} r_1 - \sum_{i\in R_1} n_i^{-1} & -\sum_{i\in R_{12}} n_i^{-1} & \cdots & -\sum_{i\in R_{1,b-1}} n_i^{-1} \\ -\sum_{i\in R_{12}} n_i^{-1} & r_2 - \sum_{i\in R_2} n_i^{-1} & \cdots & -\sum_{i\in R_{2,b-1}} n_i^{-1} \\ \vdots & \vdots & \ddots & \vdots \\ -\sum_{i\in R_{1,b-1}} n_i^{-1} & -\sum_{i\in R_{2,b-1}} n_i^{-1} & \cdots & r_{b-1} - \sum_{i\in R_{b-1}} n_i^{-1} \end{bmatrix} \tag{12}$$

is a matrix which only depends on the assignment of the design points to the blocks. In this expression, which is valid only if no design points are replicated within one block, $r_i (i = 1, 2, \dots, b)$ is the number of points in the ith block that appear more than once in the entire design. The set of these points is denoted by R_i, and the set of points the ith and the jth block have in common is denoted by $R_{ij} = R_i \cap R_j (i, j = 1, 2, \dots, b)$.

This result implies that a $\mathcal{D}$-optimal minimum support design in the presence of one fixed blocking variable can be constructed in two steps:

1. Choose the p distinct design points and replicate them as evenly as possible in order to obtain a minimum support design with n observations that maximizes $|\mathbf{X}'\mathbf{X}|$. Each of the support points then appears int(n/p) or int(n/p)+1 times in the design. Which design points are replicated most is unimportant.
2. Spread the replicated design points as evenly as possible over the blocks and avoid replicating points within a block. The assignment of the non-replicated design points to the blocks does not affect $|\mathbf{R}|$ and has therefore no impact on the $\mathcal{D}$-optimality criterion value of the experiment.

The following theorems imply that this two-step approach can be applied in situations (1) in which there is one random blocking variable, (2) in which there are two or more fixed blocking variables, and (3) in which there are two or more random blocking variables and the block sizes are equal. For the proofs of these results, we refer the reader to the Appendices 1, 2 and 3, respectively.

Theorem 1 *The determinant of the information matrix of a blocked experiment with minimum support and one blocking variable that has b levels and is treated as random is given by* $\sigma_\varepsilon^{-2p}|\mathbf{X}'\mathbf{X}|\ |\mathbf{S}|\ |\mathbf{Q}|$, *where*

$$\mathbf{S} = \begin{bmatrix} \frac{\sigma_\varepsilon^2}{\sigma_1^2} + r_1 - \sum_{i\in R_1} n_i^{-1} & -\sum_{i\in R_{12}} n_i^{-1} & \cdots & -\sum_{i\in R_{1b}} n_i^{-1} \\ -\sum_{i\in R_{12}} n_i^{-1} & \frac{\sigma_\varepsilon^2}{\sigma_1^2} + r_2 - \sum_{i\in R_2} n_i^{-1} & \cdots & -\sum_{i\in R_{2b}} n_i^{-1} \\ \vdots & \vdots & \ddots & \vdots \\ -\sum_{i\in R_{1b}} n_i^{-1} & -\sum_{i\in R_{2b}} n_i^{-1} & \cdots & \frac{\sigma_\varepsilon^2}{\sigma_1^2} + r_b - \sum_{i\in R_b} n_i^{-1} \end{bmatrix},$$

$$\mathbf{Q} = \begin{bmatrix} \frac{\eta_1}{1+k_1\eta_1} & 0 & \cdots & 0 \\ 0 & \frac{\eta_1}{1+k_2\eta_1} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \frac{\eta_1}{1+k_b\eta_1} \end{bmatrix},$$

$\eta_1 = \sigma_1^2/\sigma_\varepsilon^2$ *and* $k_1, k_2, \dots, k_b$ *represent the block sizes.*

Theorem 2 *The determinant of the information matrix of a blocked experiment with minimum support and two or more blocking variables that are treated as fixed is proportional to the product of the determinant* $|\mathbf{X}'\mathbf{X}|$ *and a determinant that depends only on the assignment of the points to the blocks.*

Theorem 3 *The determinant of the information matrix of a blocked experiment with minimum support, equal block sizes and two or more blocking variables that are treated as random is proportional to the product of the determinant* $|\mathbf{X}'\mathbf{X}|$ *and a determinant that depends only on the assignment of the points to the blocks.*

Theorems 2 and 3 show that the two-stage approach to construct $\mathcal{D}$-optimal minimum support designs can still be used when more than one blocking variable is involved in the experiment. In this context, it is important to point out that, in assigning the replicated design points to the blocks, it should be avoided that points are replicated at a given level of one of the blocking variables.

Interestingly, different designs that are equivalent in terms of $\mathcal{D}$-optimality can be obtained using the two-stage construction approach outlined here. Despite the resulting designs' equivalence in terms of the $\mathcal{D}$-optimality criterion, they may behave differently with respect to other design criteria. Any such criterion can of course be used to distinguish between alternative design options. Another consideration that can be used to rank two or more (statistically) equivalent designs is the cost of running them. Finally, it should be pointed out that efficient minimum support designs for situations with odd block sizes and random block effects, or for situations with both random and fixed block effects, can be found using the two-stage approach as well, even though we did not give a formal proof of this.

5 Mixture experiments

Mixture experiments involve blending two or more ingredients or components. In this type of experiment, the quality of the end product depends on the relative proportions of the components in the mixture. If we denote by q the number of components in the mixture and by x_i $(i = 1, 2, \ldots, q)$ the proportion contributed by the ith component, the following constraints apply to the mixture component proportions:

$$0 \leq x_i \leq 1, \tag{13}$$

and

$$\sum_{i=1}^{q} x_i = 1. \tag{14}$$

The experimental region defined by these constraints is a $(q-1)$-dimensional simplex. Often further constraints are imposed on the mixture components that result in a design region with the shape of a simplex. For example, some researchers might use lower bounds for each of the component proportions so that only experiments with mixtures containing all ingredients are performed. In such cases, it is easy to redefine the design problem for pseudocomponents, which are linear transformations of the original mixture variables, and for which the constraints

(13) and (14) apply. As zero proportions of a pseudocomponent do not necessarily correspond to a mixture where the corresponding ingredient is zero [see sections 4.2 and 4.6 of Cornell (2002)], the results presented here are also useful for researchers who wish to avoid mixtures with zero proportions for one or more components.

Popular models for modelling the response y of a mixture experiment are the canonical polynomial models introduced by Scheffé (1958). In the absence of blocking variables, the first-order Scheffé model is given by

$$y = \sum_{i=1}^{q} \beta_i x_i + \varepsilon.$$

The second-order Scheffé model is then given by

$$y = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} x_i x_j + \varepsilon.$$

A matrix notation for these models is given by (2). It is well known that $\mathcal{D}$-optimal n-point designs for these models have a minimum support, the points of which are replicated int(n/p) or int(n/p)+1 times. For the first-order Scheffé model, the $\mathcal{D}$-optimal points are the q vertices of the simplex. For the second-order model, the $q + q(q-1)/2$ $\mathcal{D}$-optimal points are the vertices and the edge midpoints, i.e. the points of a $\{q, 2\}$ simplex lattice design. Replicating these design points as evenly as possible maximizes $|\mathbf{X}'\mathbf{X}|$, which is the first step in constructing a $\mathcal{D}$-optimal minimum support design for the estimation of first- and second-order Scheffé models in the presence of blocking variables.

6 Mixture experiments in blocks

Example 1 Consider an experiment with eight runs arranged in two blocks of four observations for estimating a second-order Scheffé polynomial in three mixture variables. The determinant $|\mathbf{X}'\mathbf{X}|$ can then be maximized by choosing the six points of the $\{3, 2\}$ simplex lattice design and duplicating any two of the points. Next, one instance of each of the duplicated points is assigned to the first block and the other is assigned to the second block. Finally, two of the four non-replicated points are assigned to one block and the remaining points are assigned to the other. One possible design obtained in this way is displayed in Figure 1. This design is a $\mathcal{D}$-optimal minimum support design for the model of interest no matter whether the blocking variable is treated as random or fixed.

Example 2 Consider an experiment with 18 runs arranged in 2 blocks of 9 observations for estimating a second-order Scheffé polynomial in 4 mixture variables. To construct a $\mathcal{D}$-optimal minimum support design for this experiment in 2 blocks of size 9, 18 points have to be selected so as to maximize the determinant $|\mathbf{X}'\mathbf{X}|$. The optimal points are the ten points of the $\{4, 2\}$ simplex lattice design in four variables. Any eight of these can be duplicated. One instance of each of the duplicated points is then assigned to the first block, and the second instance is assigned

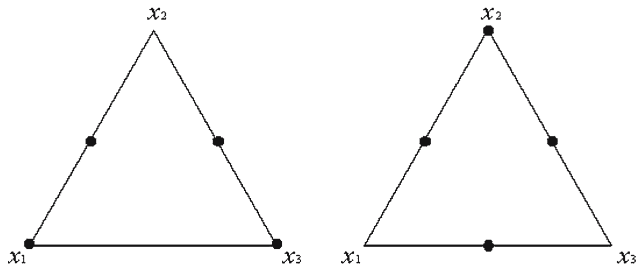

Fig. 1 $\mathcal{D}$-optimal minimum support design with two blocks of size four for the experiment described in Example 1

to the second block. Finally, one of the two remaining non-replicated design points is put in block 1 and the other is allocated to block 2. A possible design obtained in this fashion is displayed in Figure 2.

Example 3 Consider a three-component mixture experiment involving two blocking variables for estimating a second-order Scheffé polynomial model, where each blocking variable acts at two levels and two observations can be made at each combination of the levels of the blocking variables. As a result, the experiment consists of eight observations. The $\mathcal{D}$-optimal design points for a minimum support design are given by the six points of the {3, 2} simplex lattice design, two of which are duplicated. The best assignment is obtained by assigning the four duplicated points one to each block, and avoiding that any of these points occurs twice at a given level of a blocking variable. Two designs obtained in this way are given in Figure 3. The two designs are optimal for this problem in case of fixed block effects as well as in case of random block effects.

Example 4 Consider an experiment with 16 observations for estimating a second-order Scheffé model in four components and which has four blocks of size three and two blocks of size two. The blocks are generated by one fixed blocking variable with three levels and one fixed blocking variable with two levels. A $\mathcal{D}$-optimal minimum support design for this experiment is given in Figure 4. The ten distinct points of the design are the ten points of the {4, 2} simplex lattice design and thus

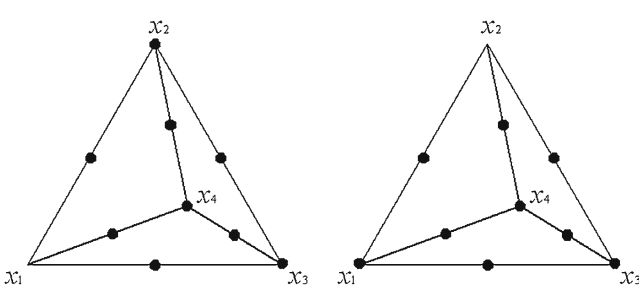

Fig. 2 $\mathcal{D}$-optimal minimum support design with two blocks of size nine for the experiment described in Example 2

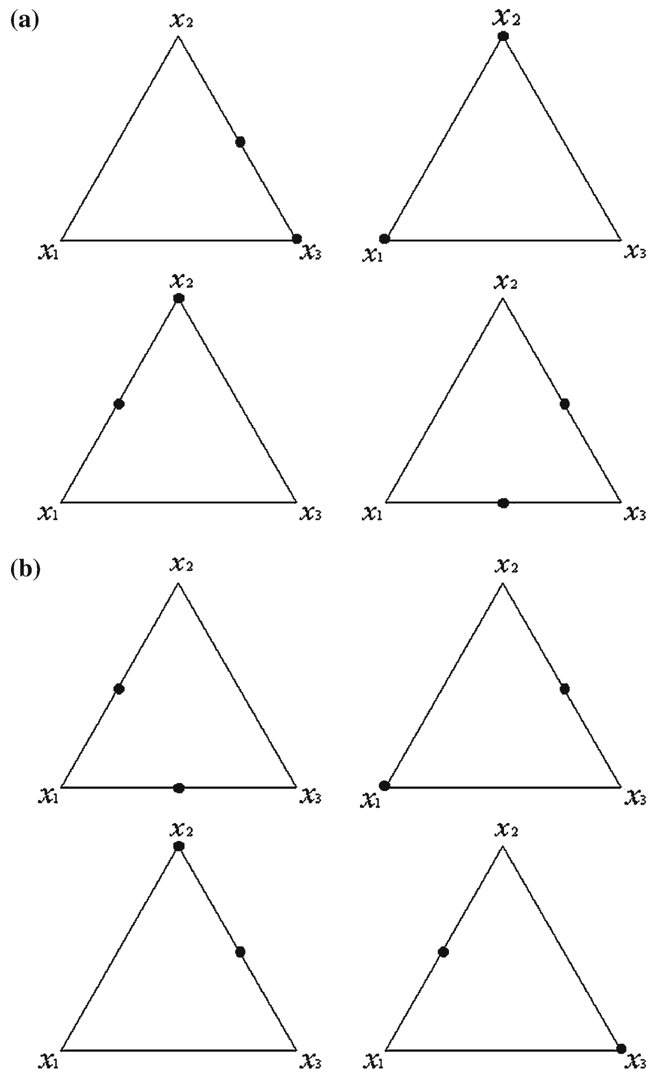

Fig. 3 Alternative $\mathcal{D}$-optimal minimum support designs for the experiment described in Example 3

maximize $|\mathbf{X}'\mathbf{X}|$. The midpoints of the six edges were duplicated in order to obtain 16 design points. The duplicated points were assigned to the blocks first so that each block contains two edge midpoints. No edge midpoints occur more than once at the same level of any of the blocking variables. Finally, a corner point is assigned to each of the blocks that should have three observations.

7 Qualitative experimental variables

The problem of designing a blocked mixture experiment with fixed block effects is similar to that of designing an experiment involving both qualitative and mixture

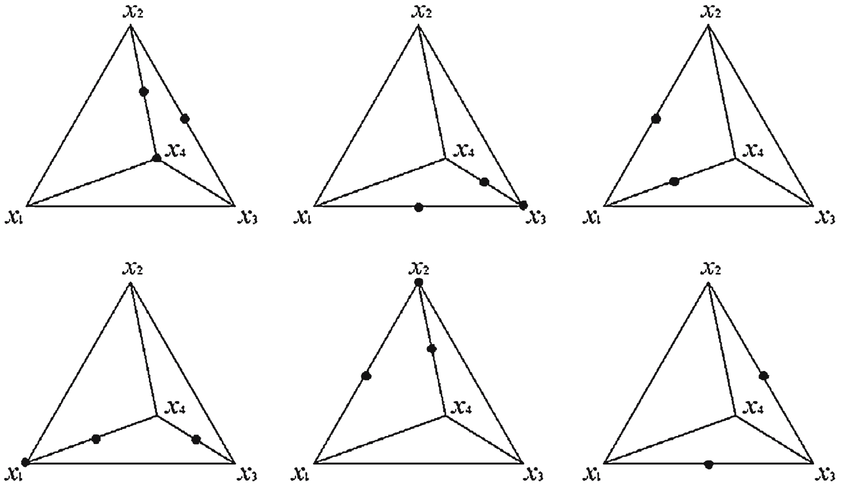

Fig. 4 $\mathcal{D}$-optimal minimum support design with two blocking variables for the experiment described in Example 4

variables. Actually, model (7) can be used for both situations. However, the effects of the blocking variables are typically considered as nuisances in a blocked mixture experiment (i.e., they are not of primary interest to the experimenter), whereas estimating the effects of qualitative variables involved in an experiment is usually as important as estimating the mixture component effects. Another difference between both problems is that in blocked experiments the numbers of runs at each combination of levels of the blocking variables are often dictated by the experimental situation whereas usually no such constraints are imposed if an experiment involves qualitative variables. It is clear that therefore other design options become available in the latter situation.

Example 5 Reconsider the minimum support designs in Figure 3. These designs are $\mathcal{D}$-optimal minimum support designs for an experiment involving three mixture components and two qualitative or blocking variables acting at two levels. It turns out, however, that the unbalanced design in Figure 5 is equally good in terms of $\mathcal{D}$-efficiency if the interest is in estimating the mixture component effects and the effects of the qualitative or blocking variables. The unbalanced design in Figure 5 and the balanced design in Figure 3b only differ in the assignment of the non-replicated design points. The optimality of the unbalanced designs is important for situations in which running experiments at certain levels of the qualitative variables is more difficult or more costly than at others.

8 Discussion

The simple design construction method presented here leads to $\mathcal{D}$-optimal designs in the class of minimum support designs, but not to $\mathcal{D}$-optimal designs in the entire

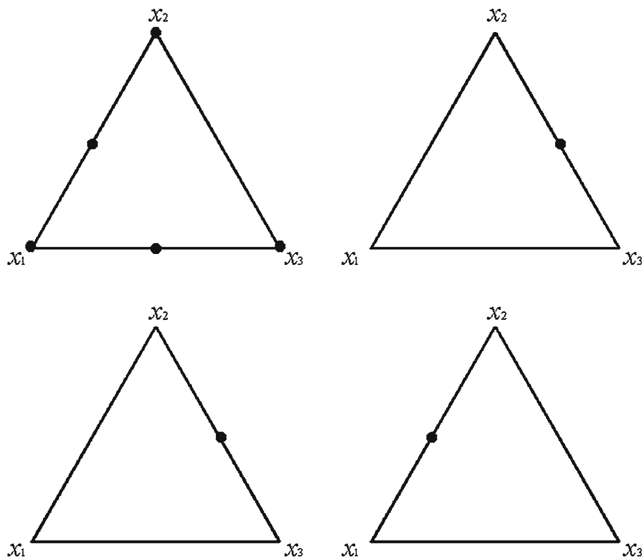

Fig. 5 Alternative $\mathcal{D}$-optimal minimum support design with two qualitative variables acting at two levels for estimating a quadratic Scheffé polynomial in three components

space of possible experimental designs. However, the $\mathcal{D}$-optimal minimum support mixture designs are highly efficient in all cases examined. This extends earlier findings for a single fixed blocking variable in Atkinson and Donev (1992) (see page 162) who indicated that moving away from minimum support designs destroys the properties that facilitate the construction of mixture designs in blocks and that this complication might yield a negligible or no increase in efficiency. The largest improvement over minimum support designs we found was for Example 3: the design in Table 1, obtained by using the algorithm of Goos and Donev (2006a), is 8.26% better in terms of $\mathcal{D}$-efficiency than the minimum support designs in Figure 3 when the blocks are treated as fixed. The candidate set used to generate the design in Table 1 was a grid of 231 equally spaced candidates on the design region. The proportions of the candidates were multiples of 0.05.

The design in Table 1 can also be found when the block effects are treated as random, for example when $\eta_1 = \eta_2 = 5$. In that case, the design is 7.32% more efficient than the minimum support designs in Figure 3. In general, the improvements that can be achieved over the minimum support designs are smaller for random

Table 1 $\mathcal{D}$-optimal design with blocks of size two for the experiment described in Example 3

		Blocking variable 1					
		Level 1			Level 2		
		x_1	x_2	x_3	x_1	x_2	x_3
Blocking variable 2	Level 1	0.70	0.30	0.00	0.00	0.00	1.00
		0.50	0.00	0.50	0.00	0.65	0.35
	Level 2	0.00	0.35	0.65	0.45	0.55	0.00
		0.00	1.00	0.00	1.00	0.00	0.00

blocks and increase with the magnitude of the variance ratios $\eta_1, \eta_2, \ldots, \eta_R$. For $\eta_1 = \eta_2 = \cdots = \eta_R = 0$, no improvements can be made over the minimum support designs.

To a large extent, the literature on blocked mixture experiments has focused on orthogonally blocked experiments. For example, Prescott (2000) proposed the orthogonally blocked design in Figure 6a for the design problem described in Example 1. This design is however far less efficient than the minimum support design displayed in Figure 1: the relative $\mathcal{D}$-efficiency of the minimum support design with respect to Prescott's orthogonally blocked design exceeds three. This implies that the minimum support design is better than three replicates of the orthogonally blocked designs in terms of $\mathcal{D}$-efficiency. Finally, it is interesting to point out that the minimum support design is only 3.11% less efficient than the $\mathcal{D}$-optimal design displayed in Figure 6b obtained using the grid of candidate points defined earlier. The precise component proportions of that design are given by (0.6, 0.4, 0), (0.5, 0, 0.5), (0, 1, 0) and (0, 0.4, 0.6) for block 1 and by (0.4, 0.6, 0), (0, 0.6, 0.4), (1, 0, 0) and (0, 0, 1) for block 2.

A drawback of the minimum support designs (and the $\mathcal{D}$-optimal designs) is that they are not orthogonally blocked. However, the minimum support designs and the $\mathcal{D}$-optimal designs perform well in terms of orthogonality, especially when the block sizes are not too small. For example, the mean efficiency factor, which measures the extent to which a blocked experiment is orthogonal [see Trinca and Gilmour (2000) for a definition of this measure], equals 83.33% for the minimum support design in Figure 1 and 90.99% for the $\mathcal{D}$-optimal design in Figure 6b. This

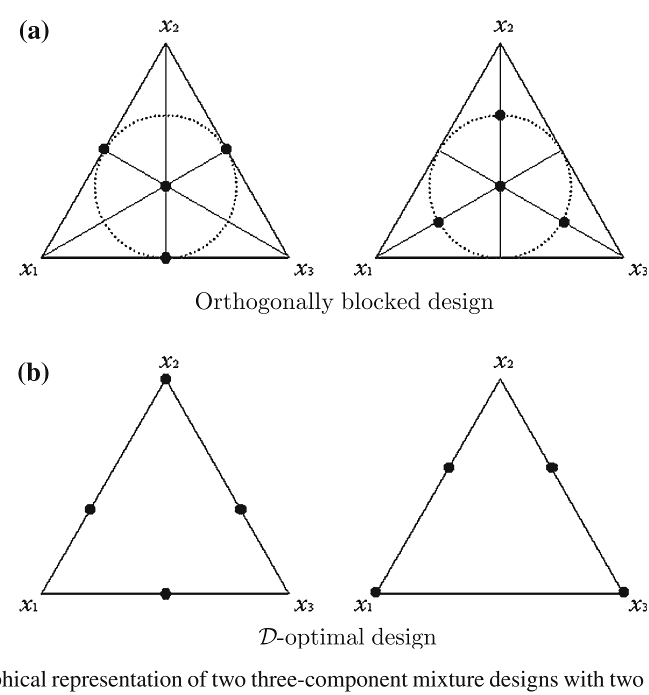

Fig. 6 Graphical representation of two three-component mixture designs with two blocks of size four for estimating a second-order Scheffé polynomial

shows that these designs are not far from being orthogonally blocked. For a more detailed discussion of the extent to which minimum support designs and $\mathcal{D}$-optimal designs are orthogonal, we refer the reader to Goos and Donev (2006b).

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Appendix 1: proof of Theorem 1

The information matrix of an experiment with one random blocking variable is

$$\begin{aligned}\mathbf{M} &= \mathbf{X}'\mathbf{V}^{-1}\mathbf{X},\\ &= \frac{1}{\sigma_\varepsilon^2}\left\{\mathbf{X}'\mathbf{X} - \left[\mathbf{X}_1'\mathbf{1}_{k_1}\ \mathbf{X}_2'\mathbf{1}_{k_2}\ \cdots\ \mathbf{X}_b'\mathbf{1}_{k_b}\right]\mathbf{Q}\begin{bmatrix}\mathbf{1}_{k_1}'\mathbf{X}_1\\ \mathbf{1}_{k_2}'\mathbf{X}_2\\ \vdots\\ \mathbf{1}_{k_b}'\mathbf{X}_b\end{bmatrix}\right\},\end{aligned} \tag{15}$$

so that

$$|\mathbf{M}| = \sigma_\varepsilon^{-2p}|\mathbf{X}'\mathbf{X}|\ |\mathbf{Q}|\ \left|\mathbf{Q}^{-1} - \begin{bmatrix}\mathbf{1}_{k_1}'\mathbf{X}_1\\ \mathbf{1}_{k_2}'\mathbf{X}_2\\ \vdots\\ \mathbf{1}_{k_b}'\mathbf{X}_b\end{bmatrix}(\mathbf{X}'\mathbf{X})^{-1}\left[\mathbf{X}_1'\mathbf{1}_{k_1}\ \mathbf{X}_2'\mathbf{1}_{k_2}\ \cdots\ \mathbf{X}_b'\mathbf{1}_{k_b}\right]\right|,$$

where

$$\mathbf{Q}^{-1} = \begin{bmatrix}\frac{\sigma_\varepsilon^2}{\sigma_1^2}+k_1 & 0 & \ldots & 0\\ 0 & \frac{\sigma_\varepsilon^2}{\sigma_1^2}+k_2 & \ldots & 0\\ \vdots & \vdots & \ddots & \vdots\\ 0 & 0 & \ldots & \frac{\sigma_\varepsilon^2}{\sigma_1^2}+k_b\end{bmatrix}.$$

Using (4), it is not difficult to see that $\mathbf{1}_{k_i}'\mathbf{X}_i(\mathbf{X}'\mathbf{X})^{-1}\mathbf{X}_i'\mathbf{1}_{k_i} = k_i - r_i + \sum_{i\in R_i} n_i^{-1}$ and that $\mathbf{1}_{k_i}'\mathbf{X}_i(\mathbf{X}'\mathbf{X})^{-1}\mathbf{X}_j'\mathbf{1}_{k_j} = \sum_{i\in R_{ij}} n_i^{-1}$ if no design points are replicated within a block. The definitions of r_i, n_i, R_i and R_{ij} can be found on the pages 3 and 5.

Appendix 2: proof of Theorem 2

First, suppose that there are two fixed blocking variables acting at b_1 and b_2 levels, respectively. Denote by $\mathbf{X}_{ij}$ the part of $\mathbf{X}$ corresponding to the ith level of the first blocking variable and the jth level of the second blocking variable, and by n_{ij} the corresponding number of runs so that $\sum_{i=1}^{b_1}\sum_{j=1}^{b_2} n_{ij} = n$. The $n\times(p+b_1+b_2-2)$ model matrix for all the fixed effects can then be written as

$$\mathbf{F} = \begin{bmatrix}
\mathbf{X}_{11} & \mathbf{1}_{n_{11}} & \mathbf{0}_{n_{11}} & \cdots & \mathbf{0}_{n_{11}} & \mathbf{1}_{n_{11}} & \mathbf{0}_{n_{11}} & \cdots & \mathbf{0}_{n_{11}} \\
\mathbf{X}_{12} & \mathbf{1}_{n_{12}} & \mathbf{0}_{n_{12}} & \cdots & \mathbf{0}_{n_{12}} & \mathbf{0}_{n_{12}} & \mathbf{1}_{n_{12}} & \cdots & \mathbf{0}_{n_{12}} \\
\vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\
\mathbf{X}_{1b_2} & \mathbf{1}_{n_{1b_2}} & \mathbf{0}_{n_{1b_2}} & \cdots & \mathbf{0}_{n_{1b_2}} & \mathbf{0}_{n_{1b_2}} & \mathbf{0}_{n_{1b_2}} & \cdots & \mathbf{0}_{n_{1b_2}} \\
\mathbf{X}_{21} & \mathbf{0}_{n_{21}} & \mathbf{1}_{n_{21}} & \cdots & \mathbf{0}_{n_{21}} & \mathbf{1}_{n_{21}} & \mathbf{0}_{n_{21}} & \cdots & \mathbf{0}_{n_{21}} \\
\mathbf{X}_{22} & \mathbf{0}_{n_{22}} & \mathbf{1}_{n_{22}} & \cdots & \mathbf{0}_{n_{22}} & \mathbf{0}_{n_{22}} & \mathbf{1}_{n_{22}} & \cdots & \mathbf{0}_{n_{22}} \\
\vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\
\mathbf{X}_{2b_2} & \mathbf{0}_{n_{2b_2}} & \mathbf{1}_{n_{2b_2}} & \cdots & \mathbf{0}_{n_{2b_2}} & \mathbf{0}_{n_{2b_2}} & \mathbf{0}_{n_{2b_2}} & \cdots & \mathbf{0}_{n_{2b_2}} \\
\vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\
\mathbf{X}_{b_1-1,1} & \mathbf{0}_{n_{b_1-1,1}} & \mathbf{0}_{n_{b_1-1,1}} & \cdots & \mathbf{1}_{n_{b_1-1,1}} & \mathbf{1}_{n_{b_1-1,1}} & \mathbf{0}_{n_{b_1-1,1}} & \cdots & \mathbf{0}_{n_{b_1-1,1}} \\
\mathbf{X}_{b_1-1,2} & \mathbf{0}_{n_{b_1-1,2}} & \mathbf{0}_{n_{b_1-1,2}} & \cdots & \mathbf{1}_{n_{b_1-1,2}} & \mathbf{0}_{n_{b_1-1,2}} & \mathbf{1}_{n_{b_1-1,2}} & \cdots & \mathbf{0}_{n_{b_1-1,2}} \\
\vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\
\mathbf{X}_{b_1-1,b_2} & \mathbf{0}_{n_{b_1-1,b_2}} & \mathbf{0}_{n_{b_1-1,b_2}} & \cdots & \mathbf{1}_{n_{b_1-1,b_2}} & \mathbf{0}_{n_{b_1-1,b_2}} & \mathbf{0}_{n_{b_1-1,b_2}} & \cdots & \mathbf{0}_{n_{b_1-1,b_2}} \\
\mathbf{X}_{b_11} & \mathbf{0}_{n_{b_11}} & \mathbf{0}_{n_{b_11}} & \cdots & \mathbf{0}_{n_{b_11}} & \mathbf{1}_{n_{b_11}} & \mathbf{0}_{n_{b_11}} & \cdots & \mathbf{0}_{n_{b_11}} \\
\mathbf{X}_{b_12} & \mathbf{0}_{n_{b_12}} & \mathbf{0}_{n_{b_12}} & \cdots & \mathbf{0}_{n_{b_12}} & \mathbf{0}_{n_{b_12}} & \mathbf{1}_{n_{b_12}} & \cdots & \mathbf{0}_{n_{b_12}} \\
\vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\
\mathbf{X}_{b_1b_2} & \mathbf{0}_{n_{b_1b_2}} & \mathbf{0}_{n_{b_1b_2}} & \cdots & \mathbf{0}_{n_{b_1b_2}} & \mathbf{0}_{n_{b_1b_2}} & \mathbf{0}_{n_{b_1b_2}} & \cdots & \mathbf{0}_{n_{b_1b_2}}
\end{bmatrix}.$$

Denoting $\mathbf{X}'_{i.} = \begin{bmatrix}\mathbf{X}'_{i1}\ \mathbf{X}'_{i2} \cdots \mathbf{X}'_{ib_2}\end{bmatrix}$, $\mathbf{X}'_{.j} = \begin{bmatrix}\mathbf{X}'_{1j}\ \mathbf{X}'_{2j} \cdots \mathbf{X}'_{b_1j}\end{bmatrix}$, and by $n_{i.}$ and $n_{.j}$ the number of rows of $\mathbf{X}_{i.}$ and $\mathbf{X}_{.j}$, the information matrix on $\boldsymbol{\xi}$ can be written as

$$\mathbf{F}'\mathbf{F} = \begin{bmatrix}
\mathbf{X}'\mathbf{X} & \mathbf{X}'_{1.}\mathbf{1}_{n_{1.}} & \cdots & \mathbf{X}'_{b_1-1,.}\mathbf{1}_{n_{b_1-1,.}} & \mathbf{X}'_{.1}\mathbf{1}_{n_{.1}} & \cdots & \mathbf{X}'_{.,b_2-1}\mathbf{1}_{n_{.,b_2-1}} \\
\mathbf{1}'_{n_{1.}}\mathbf{X}_{1.} & n_{1.} & \cdots & 0 & n_{11} & \cdots & n_{1,b_2-1} \\
\mathbf{1}'_{n_{2.}}\mathbf{X}_{2.} & 0 & \cdots & 0 & n_{21} & \cdots & n_{2,b_2-1} \\
\vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\
\mathbf{1}'_{n_{b_1-1,.}}\mathbf{X}_{b_1,1.} & 0 & \cdots & n_{b_1-1,.} & n_{b_1-1,1} & \cdots & n_{b_1-1,b_2-1} \\
\mathbf{1}'_{n_{.1}}\mathbf{X}_{.1} & n_{11} & \cdots & n_{b_1-1,1} & n_{.1} & \cdots & 0 \\
\mathbf{1}'_{n_{.2}}\mathbf{X}_{.2} & n_{12} & \cdots & n_{b_1-1,2} & 0 & \cdots & 0 \\
\vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\
\mathbf{1}'_{n_{.,b_2-1}}\mathbf{X}_{.,b_2-1} & n_{1,b_2-1} & \cdots & n_{b_1-1,b_2-1} & 0 & \cdots & n_{.,b_2-1}
\end{bmatrix},$$

the determinant of which is equal to

$$|\mathbf{F}'\mathbf{F}| = |\mathbf{X}'\mathbf{X}|\ |\mathbf{N} - \mathbf{L}(\mathbf{X}'\mathbf{X})^{-1}\mathbf{L}'|,$$

where

$$\mathbf{N} = \begin{bmatrix} n_{1.} & \cdots & 0 & n_{11} & \cdots & n_{1,b_2-1} \\ 0 & \cdots & 0 & n_{21} & \cdots & n_{2,b_2-1} \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ 0 & \cdots & n_{b_1-1,.} & n_{b_1-1,1} & \cdots & n_{b_1-1,b_2-1} \\ n_{11} & \cdots & n_{b_1-1,1} & n_{.1} & \cdots & 0 \\ n_{12} & \cdots & n_{b_1-1,2} & 0 & \cdots & 0 \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ n_{1,b_2-1} & \cdots & n_{b_1-1,b_2-1} & 0 & \cdots & n_{.,b_2-1} \end{bmatrix},$$

and

$$\mathbf{L}' = \left[\mathbf{X}'_{1.}\mathbf{1}_{n_{1.}} \;\cdots\; \mathbf{X}'_{b_1-1,.}\mathbf{1}_{n_{b_1-1,.}} \;\mathbf{X}'_{.1}\mathbf{1}_{n_{.1}} \;\cdots\; \mathbf{X}'_{.,b_2-1}\mathbf{1}_{n_{.,b_2-1}}\right].$$

Using (4), it is easy to see that the matrix product $\mathbf{L}(\mathbf{X}'\mathbf{X})^{-1}\mathbf{L}'$ only depends on the assignment of the design points to the blocks and not on their factor levels. As a result, the determinant of the information matrix can be split in two parts: one that depends on the design points and one that only depends on the assignment. A proof for more than two blocking variables that are treated as fixed can be constructed in the same way.

Appendix 3: proof of Theorem 3

Now suppose that there are R blocking variables acting at $b_1, b_2, \ldots, b_R$ levels, that the blocking variables are treated as random and that the block sizes are equal. Using the results of Goos and Donev (2006a), it can be seen that the information matrix is given by

$$\mathbf{M} = \frac{1}{\sigma_\varepsilon^2}\left\{\mathbf{X}'\mathbf{X} - \sum_{i=1}^{B_R}\sum_{j=1}^{b_i} c_i(\mathbf{X}'_{ij}\mathbf{1}_{a_i})(\mathbf{1}'_{a_i}\mathbf{X}_{ij}) + d(\mathbf{X}'\mathbf{1}_n)(\mathbf{1}'_n\mathbf{X})\right\},$$

$$= \frac{1}{\sigma_\varepsilon^2}\left\{\mathbf{X}'\mathbf{X} - \left[\mathbf{X}'_{11}\mathbf{1}_{a_1} \;\cdots\; \mathbf{X}'_{Rb_R}\mathbf{1}_{a_R} \;\mathbf{X}'\mathbf{1}_n\right]\begin{bmatrix} c_1 & 0 & \ldots & 0 \\ \vdots & \ddots & \vdots & \vdots \\ 0 & \ldots & c_R & 0 \\ 0 & \ldots & 0 & -d \end{bmatrix}\begin{bmatrix} \mathbf{X}'_{11}\mathbf{1}_{a_1} \\ \vdots \\ \mathbf{X}'_{Rb_R}\mathbf{1}_{a_R} \\ \mathbf{X}'\mathbf{1}_n \end{bmatrix}\right\},$$

where $\mathbf{X}_{ij}$ is the part of $\mathbf{X}$ assigned to the jth level of the ith blocking variable, $a_i = n/b_i$, and c_i $(i = 1, 2, \ldots, R)$ and d are constants that depend on the block size, the numbers of levels of the blocking variables and the variance components only. This matrix is similar to (15), so that its determinant can be written as a product of two determinants that are independent of each other.

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