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Optimum mixture designs under constraints on mixing components*

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Abstract

Scheffé (1958, 1963) first introduced models and designs suitable for a mixture experiment where the mean response is assumed to depend only on the relative proportions of the ingredients or components. Extensive literature on optimum designs for the estimation of parameters of different mixture models is available. The specific problem of characterization of optimal designs for estimating the optimum proportion of mixture components has been recently considered by Pal and Mandal (2006, 2008) as also by Mandal and Pal (2008) using different optimality criteria. Generalizing the work of Pal and Mandal (2006), who had dealt with the trace criterion and adopted a pseudo-Bayesian approach with invariance property of the second order moments of the optimum mixing proportions, Mandal et al. (2008) relaxed the invariance property of the second order moments of the optimum mixing proportions. In this paper, optimum designs are derived for the problem of estimating the optimum proportion of mixture components when the factor space is constrained.

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Key words: Mixture experiments; Constrained factor space; Second order models; Non-linear function; Asymptotic efficiency; Trace criterion; Optimum designs.

1 Introduction

In a mixture experiment, the response depends on the proportions $x_1, x_2, \ldots, x_q$ of q ingredients present in the mixture satisfying $x_i \geq 0, \sum_{i=1}^{q} x_i = 1$. Scheffé (1958, 1963) introduced canonical models of

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different degrees to represent the response function ζ_x. He also introduced Simplex Lattice Designs and Simplex Centroid Designs for mixture experiments. Optimality of mixture designs for the estimation of parameters of the response function was considered by Kiefer (1961), Galil and Kiefer (1977), Liu and Neudecker (1995), and others. Draper and Pukelsheim (1999) established the optimality of Weighted Centroid Designs with respect to Partial Loewner Ordering(PLO) for two and three component mixtures.

The problem of estimating the optimum mixture combination in a mixture experiment is of great practical importance. Pal and Mandal (2006) probably first attempted to find optimum designs for the estimation of optimum mixture combination. They solved the problem under the assumption that the response function can be approximated by a second degree concave function in the mixture components. The optimum mixture combination γ came out to be a non-linear function of the unknown parameters in the response function. A pseudo-Bayesian approach was pursued where a prior distribution of γ was considered with the rather restrictive assumption of invariance property of the second order moments in respect of the mixing components. The criterion used to get the optimum design was minimization of the expected trace of $MSE(\hat{\gamma})$. Some further work in this direction, using other criteria, can be found in Pal and Mandal (2008) and Mandal and Pal (2008). Since assumption of invariance on the second order moments of the components of the optimum mixture seems very restrictive, Mandal et al. (2008) solved the problem with a more general assumption on the second order moments of the prior distribution of γ.

In many practical situations, the experimenter is faced with the problem of determining the optimum mixing proportions, when certain restrictions are placed on one or more of the components. When a lower bound is specified for at least one component, the problem can be solved by introducing pseudo components (cf. Cornell,2002, pp.134). However, when an upper bound, or both lower and upper bounds, is indicated for one or more components, the problem becomes too difficult to tackle. In this case, for estimation of the parameters of the assumed model, or linear functions of the parameters, some algorithms have been proposed to find the optimum design. However, estimation of non-linear functions of parameters poses much difficulty. In this paper, we derive optimum designs for the problem with constrained factor space in cases of two and three components.

The paper is organized as follows. In Section 2, we formulate and investigate the problem. In Section 3, the optimal designs are obtained for mixtures involving two or three components.

2 Problem and the perspectives

As in Pal and Mandal (2006), we assume the response function to be quadratic concave in the components $x_1, x_2, \dots x_q$ in the factor space $\Xi = \{(x_1, x_2, \dots, x_1) | x_i \geq 0, i = 1(1)q, \sum x_i = 1\}$ and to have the form

$$\begin{aligned} E(Y|\mathbf{x}) = \zeta_x &= \sum_i \beta_{ii} x_i^2 + \sum_{i<j} \beta_{ij} x_i x_j \\ &= \mathbf{f}'(x)\beta, \end{aligned} \tag{2.1}$$

where

$$\begin{aligned} \boldsymbol{x} &= (x_1, x_2, \dots, x_q)' \\ \boldsymbol{f}(\boldsymbol{x}) &= (x_1^2, x_2^2, x_q^2, x_1, x_2, x_1 x_3, \dots, x_{q-1} x_q)' \\ \boldsymbol{\beta} &= (\beta_{11}, \beta_{22}, \dots, \beta_{qq}, \beta_{12}, \beta_{13} \dots, \beta_{q-1,q})' \end{aligned}$$

$\boldsymbol{f}(\boldsymbol{x})$ and $\boldsymbol{\beta}$ being $p \times 1$ vectors with $p = \begin{pmatrix} q+1 \\ 2 \end{pmatrix}$.

Let, $\Xi^* \subset \Xi$ denote the constrained factor space. The explicit form of Ξ^* will depend on the type of constraint imposed.

The response function (2.1) can also be expressed in the form

$$\zeta_x = \boldsymbol{x}' \boldsymbol{B} \boldsymbol{x},$$

with

$$\boldsymbol{B} = \begin{bmatrix} \beta_{11} & (1/2)\beta_{12} & (1/2)\beta_{13} \dots (1/2)\beta_{1q} \\ & \beta_{22} & (1/2)\beta_{23} \dots (1/2)\beta_{2q} \\ & \dots\dots\dots\dots\dots\dots\dots\dots\dots & \\ & & \beta_{qq} \end{bmatrix}.$$

We assume that $\boldsymbol{B}$ is negative definite and that, subject to $\sum_{i=1}^{q} x_i =$

1, ζ_x is maximized in an interior point $\boldsymbol{x} = \boldsymbol{\gamma}$ of Ξ^*, where $\boldsymbol{\gamma}$ is given by

$$\boldsymbol{\gamma} = \delta^{-1}\mathbf{B}^{-1}\mathbf{1}, \tag{2.2}$$

with $\delta = \mathbf{1}'\mathbf{B}^{-1}\mathbf{1}$. We are interested in estimating the non-linear function $\boldsymbol{\gamma}$ given by (2.2) as accurately as possible by a proper choice of a design in Ξ^*. In this paper, we shall work in the framework of *approximate* or *continuous* designs.

Let ξ be an arbitrary continuous design in Ξ and $\boldsymbol{M}(\xi, \boldsymbol{\beta}) = \int_{\Xi} f(x)f'(\boldsymbol{x})d\xi(\boldsymbol{x})$, the information matrix. For a given design ξ, we can estimate $\boldsymbol{B}$ by $\hat{\boldsymbol{B}}$, the least squares estimator of $\boldsymbol{B}$, and hence δ by $\hat{\delta}$. Then, replacing δ and $\boldsymbol{B}$ by $\hat{\delta}$ and $\hat{\boldsymbol{B}}$ respectively in (2.2), we get an estimate of $\boldsymbol{\gamma}$ as

$$\hat{\boldsymbol{\gamma}} = \hat{\delta}^{-1}\hat{\boldsymbol{B}}^{-1}\mathbf{1}. \tag{2.3}$$

Under suitable regularity assumptions on error distribution, the standard ∂-method gives, for large n, an adequate approximation of the dispersion matrix of $\hat{\gamma}$ as

$$\boldsymbol{E}\left[(\hat{\boldsymbol{\gamma}} - \boldsymbol{\gamma})(\hat{\boldsymbol{\gamma}} - \boldsymbol{\gamma})'\right] = \boldsymbol{A}(\gamma)\boldsymbol{M}^{-}(\xi, \boldsymbol{\beta})\boldsymbol{A}'(\gamma) \tag{2.4}$$

where $\boldsymbol{A}(\boldsymbol{\gamma})$ is a $q \times p$ matrix given by

$$\boldsymbol{A}(\boldsymbol{\gamma}) = \left(\frac{\partial\gamma}{\partial\beta_{11}}, \frac{\partial\gamma}{\partial\beta_{22}}, \ldots, \frac{\partial\gamma}{\partial\beta_{qq}}, \frac{\partial\gamma}{\partial\beta_{12}}, \frac{\partial\gamma}{\partial\beta_{13}}, \ldots, \frac{\partial\gamma}{\partial\beta_{q-1,q}}\right),$$

$\boldsymbol{M}(\xi, \boldsymbol{\beta})$ is the information matrix of the design ξ for the model (2.1), and $\boldsymbol{M}^{-}(\xi, \boldsymbol{\beta})$ is its generalized inverse. Here we restrict our study to the class of non-singular information matrices, as in Pal and Mandal (2006).

It has been shown in Pal and Mandal (2006) that $A(\gamma)$ can be expressed as

$$\boldsymbol{A}(\gamma) = d\begin{bmatrix} -2(q-1)\gamma_1 & 2\gamma_2 & \cdots & 2\gamma_q & \gamma_1 - (q-1)\gamma_2 & \cdots & \gamma_{q-1} + \gamma_q \\ 2\gamma_1 & -2((q-1)\gamma_2 & \cdots & 2\gamma_q & \gamma_2 - (q-1)\gamma_1 & \cdots & \gamma_{q-1} + \gamma_q \\ 2\gamma_1 & 2\gamma_2 & \cdots & 2\gamma_q & \gamma_1 + \gamma_2 & \cdots & \gamma_{q-1} + \gamma_q \\ & & \cdots & \cdots & \cdots & & \\ 2\gamma_1 & 2\gamma_2 & \cdots & 2\gamma_q & \gamma_1 + \gamma_2 & \cdots & \gamma_{q-1} + \gamma_q \\ & & \cdots & \cdots & \cdots & & \\ 2\gamma_1 & 2\gamma_2 & \cdots & 2\gamma_q & \gamma_1 + \gamma_2 & \cdots & \gamma_{q-1} - (q-1)\gamma_q \\ 2\gamma_1 & 2\gamma_2 & \cdots & -2(q-1)\gamma_q & \gamma_1 + \gamma_2 & \cdots & \gamma_q - (q-1)\gamma_{q-1} \end{bmatrix} \tag{2.5}$$

where $d = [\delta_q^{q-2}|\boldsymbol{B}|]^{-1/(q-1)}$, $|\boldsymbol{B}|$ being the determinant of the matrix $\boldsymbol{B}$.

Design optimality aims at minimizing some function of $\boldsymbol{A}(\gamma)\boldsymbol{M}^{-1}(\xi,\boldsymbol{\beta})\mathbf{A}'(\gamma)$. Since $\mathbf{A}'(\gamma)\mathbf{1}=0$, $\mathbf{A}(\gamma)\mathbf{M}^{-1}(\xi,\boldsymbol{\beta})\mathbf{A}'(\gamma)$ is singular. Hence, for comparing different designs, we consider the trace criterion

$$\phi(\boldsymbol{\gamma},\boldsymbol{M}(\xi,\boldsymbol{\beta}))=\mathbf{tr}(\mathbf{A}(\boldsymbol{\gamma})\mathbf{M}^{-1}(\xi,\boldsymbol{\beta})\mathbf{A}'(\boldsymbol{\gamma})). \tag{2.6}$$

It should be noted that the mixture model, in its canonical form, is linear in the parameters and, hence, the information matrix $\boldsymbol{M}(\xi,\boldsymbol{\beta})$ is independent of the parameters. This means that the expression in (2.6) depends on γ only through the elements of the matrix $\boldsymbol{A}(\boldsymbol{\gamma})$. Of course, this is built upon the consideration that in our search for optimal design, we may and will*disregard* the common multiplying factor d in the expression (2.5) for the matrix $\boldsymbol{A}(\boldsymbol{\gamma})$. Note that without this factor, the elements of the matrix $\boldsymbol{A}(\boldsymbol{\gamma})$ are linear in the $\boldsymbol{\gamma}$-components and, consequently, the expression in (2.6) is quadratic in the $\boldsymbol{\gamma}$-components. Therefore, assuming a prior on the first two moments of the -components is adequate. This is precisely what was done in Pal and Mandal (2006). We now continue along similar lines.

Pal and Mandal (2006), assumed a prior distribution of $\boldsymbol{\gamma}$ with $E(\gamma_i^2)=v$, $i=1,2,\ldots,q$ and $E(\gamma_i\gamma_j)=w$, $i,j=1,2,\ldots,1;i<j$ and minimized $E[\phi(\gamma,\boldsymbol{M}(\xi))]$, expectation being taken with respect to the prior distribution of $\boldsymbol{\gamma}$. Afterwards, Mandal et al. (2008) made a more general assumption on the prior moments viz.

$$E(\gamma_i^2)=v_i, i=1,2,\ldots,q \quad E(\gamma_i\gamma_j)=w_{ij}, i,j=1,2,\ldots,q; i<j. \tag{2.7}$$

Since $\sum_{i=1}^{q}\gamma_i=1,, v_i, w_{ij}$s must satisfy

$$\sum_i v_i+2\sum_{i<j}w_{ij}=1.$$

Our criterion for optimal choice of design is to minimize

$$\phi(\xi)=E\phi(\gamma,\boldsymbol{M}(\xi,\boldsymbol{\beta}))=\mathrm{tr}(\boldsymbol{M}^{-1}(\xi,\boldsymbol{\beta})E(\boldsymbol{A}'(\gamma)\boldsymbol{A}(\gamma))). \tag{2.8}$$

3 Optimum designs

Here we find optimum designs for $q=2,3$.

3.1 Case of two components

Let us consider $\Xi^* = \{\boldsymbol{x} | x_1, x_2 \geq 0, x_1 \leq c, x_1 + x_2 = 1\}$.

Since $x_1 + x_2 = 1$, invoking the result of Liski et al. (2002) , and arguing as in Mandal et al. (2008), for finding the optimum design we may restrict ourselves to the class of three-point designs with support points $(0,1), (c, 1-c)$ and $(d_1, 1-d_1)$, $d_1 \in (0, c)$. Let η denote a three-point design with masses α_1, α_2 and $1 - \alpha_1 - \alpha_2$, respectively, at the support points $(0,1), (c, 1-c)$ and $(d_1, 1-d_1)$, $d_1 \in (0, c)$.

The moment matrix of the design is

$$M(\eta) = \begin{pmatrix} a_1 & b & c_1 \\ & a_2 & c_2 \\ & & b \end{pmatrix} \tag{3.1}$$

where

$$\begin{aligned} a_1 &= c^4\alpha_1 + d_1^4(1-\alpha_1-\alpha_2) \\ a_2 &= (1-c)^4\alpha_1 + (1-d_1)^4(1-\alpha_1-\alpha_2) \\ c_1 &= c^3(1-c)\alpha_1 + d_1^3(1-d_1)(1-\alpha_1-\alpha_2) \\ c_2 &= c(1-c)^3\alpha_1 + d_1(1-d_1)^3(1-\alpha_1-\alpha_2) \\ b &= c^2(1-c)^2\alpha_1 + d_1^2(1-d_1)^2(1-\alpha_1-\alpha_2). \end{aligned} \tag{3.2}$$

An alternative representation of model (2.1) in the two component case is

$$\zeta_x = \theta_{11}x_1(x_1-d_1) + \theta_{22}(x_2-(1-c))(x_2-(1-d_1)) + \theta_{12}(x_1-c)(x_2-1) \tag{3.3}$$

where $\theta = (\theta_{11}, \theta_{22}, \theta_{12})$ and $\beta = (\beta_{11}, \beta_{22}, \beta_{12})$ are related by

$$\beta = L\theta$$

with

$$L = \begin{pmatrix} 1-d_1 & (1-c)(1-d_1) & -(1-c) \\ 0 & cd_1 & 0 \\ -d_1 & -d_1(1-c) - c(1-d_1) & c \end{pmatrix}.$$

Then,

$$M(\xi,\theta)=\begin{pmatrix}\alpha_2c^2(c-d_1)^2 & 0 & 0\\ & \alpha_1c^2d_1^2 & 0\\ & & (1-\alpha)d_1^2(c-d_1)^2\end{pmatrix} \tag{3.4}$$

where $\alpha=\alpha_1+\alpha_2$.

Hence,

$$M^{-1}(\xi,\beta)=LM^{-1}(\xi,\theta)L' \tag{3.5}$$

Therefore, we get

$$\phi(\xi)=\ \mathrm{tr}\, M^{-1}(\xi,\theta)G,$$

where $G=((g_{ij}))=L'E(A'(\gamma)A(\gamma))L$

$$\begin{aligned} g_{11} &= 8(1-d_1)^2v_1+2d_1^2(v_1+v_2-2w_{12})-8d_1(1-d_1)(w_{12}-v_1)\\ g_{22} &= 8(1-c)^2(1-d_1)^2v_1+8c^2d_1^2v_2+2(d_1(1-c)+c(1-d_1))^2(v_1+v_2-2w_{12})\\ &\quad -16c(1-c)d_1(1-d_1)w_{12}-8(1-c)(1-d_1)(d_1(1-c)+c(1-d_1))(w_{12}-v_1)\\ &\quad -8cd_1(d_1(1-c)+c(1-d_1))(w_{12}-v_2)\\ g_{33} &= 8(1-c)^2v_1+2c^2(v_1+v_2-2w_{12})-8c(1-c)(w_{12}-v_1). \end{aligned}$$

Thus, for given d_1,

$$\phi(\xi)=\frac{g_{11}}{\alpha_2c^2(c-d_1)^2}+\frac{g_{22}}{\alpha_1c^2d_1^2}+\frac{g_{33}}{(1-\alpha)d_1^2(c-d_1)^2}\geq\left(\sum_i\sqrt{g_{ii}^*}\right)^2, \tag{3.6}$$

where

$$g_{11}^*=\frac{g_{22}}{c^2d_1^2},\quad g_{22}^*=\frac{g_{11}}{c^2(c-d_1)^2},\quad g_{33}^*=\frac{g_{33}}{d_1^2(c-d_1)^2}, \tag{3.7}$$

with equality in (3.6) holding for

$$\alpha_i=\alpha_i^*(d_1)=\frac{\sqrt{g_{ii}^*}}{\sum_j\sqrt{g_{jj}^*}},i=1,2. \tag{3.8}$$

Suppose d_1^* is the value of d_1 minimizing $\left(\sum_i\sqrt{g_{ii}^*}\right)^2$. Then the optimal design assigns masses $\alpha_1^*(d_1^*),\alpha_2^*(d_1^*)$ and $1-\alpha_1^*(d_1^*)-\alpha_2^*(d_1^*)$, respectively, at the support points $(0,1),(c,1-c)$ and $(d_1^*,1-d_1^*)$.

Table 3.1: Showing optimum designs and the minimum trace for different combinations of v_1, v_2, w_{12} and c

v_1	v_2	w_{12}	c	d_1^*	α_2^*	α_1^*	Min. Trace
0.26	0.26	0.24	1	0.5	0.3646	0.3646	17.4530
			0.8	0.3501	0.4127	0.1950	41.9121
			0.6	0.2934	0.3894	0.1256	210.158
			0.4	0.1998	0.3248	0.1765	2010.959
0.26	0.30	0.22	1	0.5092	0.3029	0.3337	29.0988
			0.8	0.3824	0.3514	0.2338	65.6210
			0.6	0.2942	0.3520	0.1710	269.3375
			0.4	0.1996	0.3148	0.1886	2189.214
0.30	0.28	0.21	1	0.4967	0.3144	0.3010	34.6172
			0.8	0.3856	0.3470	0.2151	84.9551
			0.6	0.2963	0.3403	0.1765	355.406
			0.4	0.1998	0.3071	0.1956	2750.812
0.30	0.30	0.20	1	0.5	0.3000	0.3000	40.0000
			0.8	0.3895	0.3326	0.2250	95.9710
			0.6	0.2966	0.3311	0.1866	389.2969
			0.4	0.1997	0.3038	0.1998	2837.252
0.30	0.40	0.15	1	0.5048	0.2435	0.3008	66.0020
			0.8	0.3976	0.2694	0.2636	148.5268
			0.6	0.2982	0.2835	0.2334	510.6013
			0.4	0.1997	0.2900	0.2148	3262.091
0.30	0.48	0.11	1	0.5052	0.2435	0.3008	86.3329
			0.8	0.3997	0.2694	0.2636	189.1602
			0.6	0.2990	0.2835	0.2334	609.3615
			0.4	0.1998	0.2812	0.2240	3595.568
0.40	0.30	0.15	1	0.4950	0.2997	0.2598	66.0020
			0.8	0.3949	0.3146	0.2165	170.6315
			0.6	0.2986	0.3092	0.2016	660.2805
			0.5	0.2494	0.3006	0.2049	1570.399
0.40	0.40	0.10	1	0.5000	0.2717	0.2717	92.1051
			0.8	0.3978	0.2887	0.2378	223.0933
			0.6	0.2990	0.2918	0.2196	786.0946
			0.5	0.2496	0.2882	0.2183	1786.614
0.48	0.30	0.11	1	0.4948	0.3008	0.2435	86.3329
			0.8	0.3965	0.3086	0.2136	230.0149
			0.6	0.2991	0.3012	0.2068	881.3037
			0.5	0.2496	0.2936	0.2106	2062.470
0.48	0.48	0.02	1	0.5000	0.2650	0.2650	133.2798
			0.8	0.3989	0.2773	0.2413	323.6493
			0.6	0.2994	0.2803	0.2284	1104.629
			0.5	0.2498	0.2782	0.2270	2446.772

Remark 3.1 *From Table 3.1, the following observations have been made:*
(a) For fixed v_1, v_2 and w_{12}, (i) min. trace $\downarrow$ c; (ii) $d_1 \uparrow c$; (iii) $1-\alpha_1-\alpha_2$ (mass at d_1) $\downarrow c$. (b) For fixed $v_2(v_1)$ and c, min trace $\uparrow v_1(v_2)$.

3.2 Case of three components

Here the model is

$$E(Y|x) = \zeta_x = \sum_{i=1}^{3} \beta_{ii} x_i^2 + \sum_{\substack{i,j=1\\ i<j}}^{3} \beta_{ij} x_i x_j. \tag{3.9}$$

We shall assume that

$$v_1 = v_2, w_{13} = w_{23}. \tag{3.10}$$

For simplicity, we write model (3.9) as

$$\zeta_x = \beta_{11}x_1^2 + \beta_{22}x_2^2 + \beta_{12}x_1x_2 + \beta_{33}x_3^2 + \beta_{13}x_1x_3 + \beta_{23}x_2x_3.$$

Then, writing $\boldsymbol{A}(\gamma) = \left(\frac{\partial\gamma}{\partial\beta_{11}}, \frac{\partial\gamma}{\partial\beta_{22}}, \frac{\partial\gamma}{\partial\beta_{12}}, \frac{\partial\gamma}{\partial\beta_{33}}, \frac{\partial\gamma}{\partial\beta_{13}}, \frac{\partial\gamma}{\partial\beta_{23}}\right)$, we have

$$\begin{aligned}
&\boldsymbol{E}(\boldsymbol{A}'(\gamma)\boldsymbol{A}(\gamma))\\
&= d^2 \begin{bmatrix}
24v_1 & -12w_{12} & -6(v_1-2w_{12}) & -12w_{13} & -6(v_1-2w_{13}) & -6(w_{12}+w_{13}) \\
 & 24v_1 & -6(v_1-2w_{12}) & -12w_{13} & -6(w_{12}+w_{13}) & -6(v_1-2w_{13}) \\
 & & 6(2v_1-w_{12}) & -12w_{13} & -3(v_1+w_{12}-w_{13}) & -3(v_1+w_{12}-w_{13}) \\
 & & & 24v_3 & -6(v_3-2w_{13}) & -6(v_3-2w_{13}) \\
 & & & & 6(v_1+v_3-w_{13}) & -3(v_3+2w_{13}-2w_{12}) \\
 & & & & & 6(v_1+v_3-w_{13})
\end{bmatrix},
\end{aligned}$$

where $d = [3\delta|B|]^{-1/2}$.

It may be noted that in many practical situations the mixing proportions are constrained, like say, a particular ingredient may be essential to be present in the mixture in at least or at most a certain proportion. In our study, let us assume that $0 \le x_3 \le c$, where $0 < c \le 1$. To obtain the optimum design in such a situation, we proceed as follows.

3.2.1 A Heuristic Search for Optimum Design

We note that under the assumption (3.10), the problem is invariant with respect to the first two components and hence the optimum design must also have the same invariance property with respect to the first two components. Now arguing as in Mandal et al. (2008), the response function can be represented as a quadratic in x_3 so that for the optimum design x_3 will take the three distinct values $0, c$ and some $a \in (0, c)$. In view of the invariant case considered in Pal and Mandal (2006) or the non-invariant case of Mandal et al. (2008), we restrict to the following class of designs with support points as below:

<table>
<tr><th>x_1</th><th>x_2</th><th>weight</th><th>x_3</th><th>weight</th></tr>
<tr><td>1</td><td>0</td><td>α</td><td rowspan="3">0</td><td rowspan="3">W_1</td></tr>
<tr><td>0</td><td>1</td><td>α</td></tr>
<tr><td>1/2</td><td>1/2</td><td>$1-2\alpha$</td></tr>
<tr><td>$(1-c)/2$</td><td>$(1-c)/2$</td><td>1</td><td>c</td><td>W_2</td></tr>
<tr><td>$1-a$</td><td>0</td><td>1/2</td><td rowspan="2">a</td><td rowspan="2">W_3</td></tr>
<tr><td>0</td><td>$1-a$</td><td>1/2</td></tr>
</table>

where $0 \le \alpha \le 1$, $a \in (0, c)$, $W_i \ge 0$, $i = 1, 2, 3$, $W_1 + W_2 + W_3 = 1$. Here W_1, W_2 and W_3 denote the weights attached to $x_3 = 0, c$ and a, respectively, while the third column gives the weights for different (x_1, x_2) combinations when x_3 is given.

Let us denote such a design as $\xi(\alpha, a, \boldsymbol{W})$. Then, after a little algebra, the information matrix for the design comes out to be

$$M(\xi) = \boldsymbol{D\Lambda D}',$$

where

$$\begin{aligned}
\boldsymbol{D} &= \begin{bmatrix} \sqrt{\alpha} & 0 & b & \frac{(1-c)^2}{4} & \frac{(1-a)^2}{\sqrt{2}} & 0 \\ 0 & \sqrt{\alpha} & b & \frac{(1-c)^2}{4} & 0 & \frac{(1-a)^2}{\sqrt{2}} \\ 0 & 0 & b & \frac{(1-c)^2}{4} & 0 & 0 \\ 0 & 0 & 0 & c^2 & \frac{a^2}{\sqrt{2}} & \frac{a^2}{\sqrt{2}} \\ 0 & 0 & 0 & \frac{c(1-c)}{2} & \frac{a(1-a)}{\sqrt{2}} & 0 \\ 0 & 0 & 0 & \frac{c(1-c)}{2} & 0 & \frac{a(1-a)}{\sqrt{2}} \end{bmatrix}, \\
\boldsymbol{\Lambda} &= \mathrm{Diag}(\boldsymbol{W}_1\boldsymbol{I}_3, W_2, W_3\boldsymbol{I}_2), b = \sqrt{1-2\alpha}\,\frac{1}{4}.
\end{aligned}$$

Therefore, for the design ξ, the criterion function (2.8) reduces to

$$\begin{aligned}\phi(\xi(\alpha, a, \boldsymbol{W})) &= \operatorname{tr}\Lambda^{-1}\left[D^{-1}\boldsymbol{E}(\boldsymbol{A}'(\gamma)A(\gamma))D'^{-1}\right]\\ &= \frac{g^*_{11}}{W_1}+\frac{g^*_{22}}{W_2}+\frac{g^*_{33}}{W_3}\end{aligned}$$

where

$$D^{-1}E(A'(\gamma)A(\gamma))D'^{-1} = ((g_{ij}))$$

$$\begin{aligned}
g^*_{11} &= g_{11}+g_{22}+g_{33},\, g^*_{22} = g_{44},\, g^*_{33} = g_{55}+g_{66},\\
g_{11} &= g_{22} = \frac{1}{\alpha}\Big[24v_1 + 6(4v_1 - 5w_{12}) + 6(v_1 - w_{12} - 3w_{13})\frac{1-a}{a}\\
&\quad +6(v_1+v_3-w_{13})\left(\frac{1-a}{a}\right)^2\left[\frac{c(1-a)}{(c-a)}\right]^2\\
&\quad +6\frac{(1-a)^2}{a^2}\ \frac{(1-c)}{4(c-a)^2}\left\{4v_3(1-a)^2(1-c)+2(v_3-2w_{13})(1-a)c(2-a)\right.\\
&\quad \left.-(v_3+2w_{13}-2w_{12})a(2c-a-ac)\right\}\Big]\\
&= \frac{g^*}{2\alpha},\ \text{say}\\
g_{33} &= \frac{96}{1-2\alpha}\left[(2v_1-w_{12})+\frac{(1-c_2)^2(1-a)}{c_2(c_2-a)}\left\{w_{13}-(v_1+w_{12}-w_{13})\frac{a}{2(1-a)}\right\}\right.\\
&\quad \left.+\frac{(1-c_2)^4(1-a)^2}{c_2^2(c_2-a)}\left\{v_3+(v_3-2w_{13})\frac{a}{1-a}+(2v_1-v_3-4w_{13}+2w_{12})\left(\frac{a}{(1-a)}\right)^2\right\}\right]\\
&= \frac{h^*}{1-2\alpha},\ \text{say}\\
g_{44} &= \frac{6(1-a)^2}{c_2^2(c-a)^2}\left[4v_3+4(v_3-2w_{13})\frac{a}{1-a}+(2v_1+v_3-4w_{13}+2w_{12})\left(\frac{a}{1-a}\right)^2\right]\\
g_{55} &= g_{66} = \frac{3}{a^2(c-a)^2}\left[(v_1+v_3-w_{13})\left(\frac{2c-a-ac}{1-a}\right)^2+\right.\\
&\quad (1-c)^2\left\{4v_3+2(v_3-2w_{13})\frac{a}{1-a}+(v_1+v_3-w_{13})\left(\frac{a}{1-a}\right)^2\right\}\\
&\quad \left.+(1-c)\frac{2c-a-ac}{1-a}\left\{2(v_3-2w_{13})-(v_3+2w_{13}-2w_{12})\frac{a}{1-a}\right\}\right].
\end{aligned}$$

For given a and $\boldsymbol{W}$, $\phi(\xi(\alpha, a, \boldsymbol{W}))$ is minimized at $\alpha = \alpha_0 = \frac{\sqrt{g^*}}{2\sqrt{g^*}+\sqrt{2h^*}}$.

Then, at $\alpha = \alpha_0$,

$$\phi(\xi(\alpha_0, a, \boldsymbol{W})) = \phi(\xi(a, \boldsymbol{W})) = \frac{g^*_{11,0}}{W_0}+\frac{g^*_{22,0}}{W_1}+\frac{g^*_{33,0}}{W_a} \geq \left(\sum_i \sqrt{g^*_{ii,}}\right)^2 \tag{3.11}$$

where $g^*_{ii,0} = g^*_{ii}\big|_{\alpha=\alpha_0}, i = 1, 2, 3.$

Equality holds in (3.11) at $W_i = W_i(a) = \frac{\sqrt{g^*_{ii,0}}}{\sum_i \sqrt{g^*_{ii,0}}}$ $i = 1, 2, 3.$

Hence, given a,

$$\phi(\xi(\alpha, a, \boldsymbol{W})) \geq \phi(\xi(a, \boldsymbol{W}(a)) = \left(\sum_i \sqrt{g^*_{ii,0}}\right)^2 \quad \text{for all} \quad \alpha, \boldsymbol{W}. \tag{3.12}$$

We now find optimal a which minimizes the R.H.S. of (3.12). As algebraic deduction of optimal a is intractable, we have indicated the optimal value a^* of a and hence those of α and $\boldsymbol{W}$, for some combinations $(v_1 = v_2, v_3, w_{12}, w_{13} = w_{23})$ between w_{12} and w_{13} in Table 3.2. It may be noted that this design is optimal within the class of designs.

$$D_0 = \{\xi(\alpha, a, \boldsymbol{W});\ 0 \leq \alpha \leq 1,\ 0 < a < 1,\ W_i \geq 0,\ i = 1, 2, 3,\ W_1 + W_2 + W_3 = 1\} \tag{3.13}$$

Table 3.2 Showing optimal values of a, α and $\boldsymbol{W}$ for different combinations of $(v_1 = v_2, v_3, w_{12}, w_{13} = w_{23})$ and c.

	Parameters				Optimal			Trace
c	$v_1 = v_2$	v_3	w_{12}	$w_{13} = w_{23}$	a	α	W_1 W_2 W_3	
0.99	0.20	0.1	0.150	0.0500	0.4833	0.2520	0.4455 0.1095 0.4450	491.4126
	0.20	0.1	0.120	0.0650	0.4866	0.2512	0.4714 0.0967 0.4319	488.6627
	0.20	0.1	0.100	0.0750	0.4905	0.2506	0.4890 0.0872 0.4238	484.5852
	0.15	0.2	0.120	0.0650	0.4960	0.2508	0.3793 0.1424 0.4783	483.7883
	0.10	0.2	0.065	0.1175	0.5082	0.2210	0.3802 0.1263 0.4935	287.8529
0.9	0.20	0.1	0.150	0.0500	0.4526	0.2586	0.4282 0.1267 0.4451	559.3978
	0.20	0.1	0.120	0.0650	0.4579	0.2558	0.4549 0.1146 0.4305	543.4693
	0.20	0.1	0.100	0.0750	0.4636	0.2541	0.4736 0.1054 0.4210	530.1226
	0.15	0.2	0.120	0.0650	0.4668	0.2594	0.3622 0.1671 0.4707	567.9032
	0.10	0.2	0.065	0.1175	0.4920	0.2213	0.3649 0.1617 0.4734	316.4064
0.85	0.20	0.1	0.150	0.0500	0.4334	0.2640	0.4176 0.1373 0.4451	612.0382
	0.20	0.1	0.120	0.0650	0.4394	0.2595	0.4443 0.1260 0.4297	586.1483
	0.20	0.1	0.100	0.0750	0.4459	0.2568	0.4734 0.1172 0.4194	565.8259
	0.15	0.2	0.120	0.0650	0.4669	0.2666	0.3622 0.1812 0.4667	637.7346
	0.10	0.2	0.065	0.1175	0.4714	0.2220	0.3541 0.1854 0.4605	343.3944

In the following subsection we examine the optimality of the design $\xi(a^*, \boldsymbol{W}(a^*))$ within the entire class.

3.2.2 Optimality or Non-optimality of $\xi(a^*, \boldsymbol{W}(a^*))$

To verify the optimality of $\xi(a^*, \boldsymbol{W}(a^*))$ in the entire class, we use the equivalence theorem of Kiefer (1974) which, for the given problem, can be stated as follows (cf. Pal and Mandal (2007)):

Theorem 3.1 *A necessary and sufficient condition for a mixture designc ξ to be optimum is that*

$$f(x)'\boldsymbol{M}^1(\xi)(\boldsymbol{E}(\boldsymbol{A}'(\gamma)\boldsymbol{A}(\gamma)))\boldsymbol{M}^1(\xi)f(x) \leq tr\,\boldsymbol{M}^1(\xi)(E(\boldsymbol{A}'(\gamma)\boldsymbol{A}(\gamma))) \tag{3.14}$$

holds for all x in the factor space Ξ^*.

Equality in (3.14) holds at the support points of ξ.

We have checked the above condition by taking several combinations of $c, v_i, w_{ij}; i, j = 1, 2, 3$. It has been seen that when $c = 1$, condition (3.14) is satisfied for all x in Ξ^*, so that the design is optimum, as was observed by Mandal et al (2008). However, when c takes some value les than one, condition (3.14) is satisfied at all points except for a very small area in Ξ^*. Hence the design $\xi(a^*, \boldsymbol{W}(a^*))$ is not optimum in the entire class. A closer look at the following table shows that the condition (3.14) is violated at some of the support points of $\xi(a^*, \boldsymbol{W}(a^*))$ which indicates that more weights are to be attached at those support points.

Table 3.3: Showing the upper bound c^* of x_3 so that the equivalence theorem is satisfied at all points on the plane $x_1 = 0$ or $x_2 = 0$ for the design $\xi(a^*, \boldsymbol{W}(a^*))$ for given $(v_1 = v_2, v_3, w_{12}, w_{13} = w_{23}, c)$.

$v_1 = v_2$	w_{12}	$w_{13} = w_{23}$	v_3	c^*			
				$c = 0.9$	$c = 0.8$	$c = 0.7$	$c = 0.5$
0.2	0.15	0.05	0.1	0.8947	0.7844	0.6743	0.4602
0.15	0.2	0.12	0.065	0.8913	0.7759	0.6642	0.4565
0.1	0.2	0.065	0.1175	0.8742	0.7263	0.6104	0.4399

The above table shows that for given $(v_1 = v_2, v_3, w_{12}, w_{13} = w_{23})$ as c decreases, c^* deviates more from it. This indicates increase in the region of violation of the Equivalence Theorem with decrease in c.

To find optimum design sequentially one can start with this design $\xi(a^*, \boldsymbol{W}(a^*))$ and use any of the standard algorithms, like V algorithm of Federov(1972), to reach the optimum. Since, however, we are interested in determining/recommending single-step design, we have considered some competitors of $\xi(a^*, \boldsymbol{W}(a^*))$ and compared their performances for different combinations of the given parameters of the design in the next section.

4 A competitive design

In the last section we have observed that the design $\xi(a^*, \boldsymbol{W}(a^*))$ is not optimum in the entire class of competing designs. In this section we propose another competitive design $\xi_1(a_1, \boldsymbol{W}(a_1))$, which seems to be a strong contender for the target design:

x_1	x_2	weight	x_3	weight
1	0	α	0	W_1
0	1	α		
1/2	1/2	$1-2\alpha$		
$1-c$	0	1/2	c	W_2
0	$1-c$	1/2		
$1-a_1$	0	1/2	a_1	W_3
0	$1-a_1$	1/2		

where $0 \leq \alpha \leq 1$, $a_1 \in (0, c)$, $W_i \geq 0$, $i = 1, 2, 3$, $W_1 + W_2 + W_3 = 1$ and W_i s are defined as before.

For the design $\xi_1(a_1, \boldsymbol{W}(a_1))$, we use the same weights α, W_1, W_2 and W_3 as in the optimum design for the unconstrained factor space, considered by Mandal et al (2008), with the mass W_2, assigned to the point $(0, 0, 1)$, divided equally among the two points $(1-c, 0, c)$ and $(0, 1-c, c)$. The optimum a_1, denoted by a_1^* is determined by minimizing the criterion function. The relative performance of the two designs is given in Table 4.1.

Table 4.1: Showing the constrained design $\xi_1(a_1^*, \boldsymbol{W}(a_1^*))$ and its comparison with the constrained design $\xi(a^*, \boldsymbol{W}(a^*))$

Parameters				**Values of**		c	$\xi_1(a_1^*, \boldsymbol{W}(a_1^*))$		**Trace of** $\xi(a^*, \boldsymbol{W}(a^*))$
$v_1 = v_2$	v_3	w_{12}	$w_{13} = w_{23}$	α	W_1 / W_2 / W_3		a_1^*	**Trace**	
0.1	0.2	0.065	0.1175	0.2210	0.3818	0.99	0.5082	287.8265	287.8529
						0.9	0.4855	315.8616	316.4064
					0.1229	0.85	0.4609	344.8327	343.3944
						0.7	0.3506	578.6438	554.3565
					0.4953	0.5	0.2274	2395.1697	2438.563
0.15	0.2	0.12	0.065	0.2501	0.4897	0.99	0.4954	483.7396	483.7883
						0.9	0.4577	564.5837	567.9032
					0.0863	0.85	0.4319	631.9667	637.7346
						0.7	0.3465	1054.5752	1066.964
					0.4240	0.5	0.2379	3639.1909	3836.257
0.2	0.1	0.15	0.05	0.2514	0.4473	0.99	0.4827	491.3899	491.4126
						0.9	0.4448	558.0290	559.3978
					0.1077	0.85	0.4211	610.2784	612.0382
						0.7	0.3424	904.2737	955.737
					0.4450	0.5	0.2346	2464.5404	2429.956

5 Conclusion

In this paper we have attempted to obtain the optimum design for the estimation of optimum proportion γ in a mixture experiment, when the factor space is constrained. For the case of two components, the design obtained is found to be optimum in the entire class. For the case of three components, the optimum design within a class of six-point designs, defined in subsection 3.2.1, has been derived. However, using Equivalence Theorem, it is observed that the design is not optimum in the entire class of competing designs. Numerically, it is seen that the Equivalence Theorem is particularly violated at the two boundary points $(1-c, 0, c)$ and $(0, 1-c, c)$. This indicates that some mass should be allotted to these points. In view of that, in Section 4, we have introduced another class of seven-point designs, where the mass at the point $(0, 0, 1)$ in the optimum unconstrained design has been distributed equally to the two boundary points $(1-c, 0, c)$ and $(0, 1-c, c)$. By taking several combinations of the parameters, we have compared the performance of the two designs. It is observed that, in terms of the criterion function, the two are very close to one other. So, starting with any one of these designs, one may now use a standard numerical algorithm to reach the optimum design.

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