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Optimal Designs for Estimation of Optimum Mixtures and Optimum Amount in a Multiresponse Mixture Experiment

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Abstract

Optimal designs for estimating the parameters and also the optimum factor combinations in multi-response experiments have been considered by some authors. However, the existing literature on mixture experiments shows studies mainly in the single response case. In this paper an attempt has been made to investigate optimum designs for estimating optimum mixing proportions and also the optimum amount of mixture in a multi-response experiment. The pseudo-Bayesian approach has been used, and the support points of the optimum design are found to be the union of the support points of a weighted centroid design and a three-point symmetric design, with support points at the two extremes and one at the centre.

Keywords: Multiple responses, mixture experiment, mixture-amount model, linear optimality criterion, optimal design, invariance, weighted centroid design.

1. Introduction

Design of experiments has vast application in different fields, such as engineering, pharmaceutical, biomedical, environmental and epidemiological research. In these areas, it is often necessary to measure more than one response for each setting of control variables. Such experiments are called multi-response experiments. The responses may be correlated so that they cannot be studied independently. In other cases, the cost of experimenting and collecting data using single response experiments makes one reconsider the problem as a multi-response design of experiments. Roy et al. [1] first developed techniques for multi-response experiments. Fedorov [2] established a theoretical foundation and developed a recursive algorithm for generating multi-response approximate D-optimal designs. Chang [3] studied the properties of D-optimal designs for multi-response models and proved that the optimal design of a multi-response model whose response functions have the same forms coincide with that of a single response model of the same form. Krafft and Schaefer [4] considered a linear regression model with a one-dimensional control variable and an m-dimensional response variable. Bischoff [5] found special conditions under which D-optimal designs are optimal for problems with correlated observations and extended this finding to special multi- response models. Imhof [6] extended the first-order model of Krafft and Schaefer [4] to a second-order model. Chang et al. [7] generated D-optimal designs for a simple m-dimensional response model with a single control variable. Mandal [8] investigated D-optimal designs for the estimation of the optimum factor combinations in a multi-response experiment.

In mixture experiments, optimum designs for parameter estimation have been considered by several authors like Kiefer [9], Atwood [10], Galil and Kiefer [11], Liu and Neudecker [12], to name a few. For estimation of the optimum composition of a mixture, optimum designs have been investigated by Pal and Mandal [13-15], Mandal and Pal [16], among others. All these studies relate to mixture experiments with single response. However, there are many practical situations where the experimenter is interested in more than one characteristic of the output. For example, in pharmaceutical or biomedical research, though the efficacy of a drug or remedy is of primary concern, one cannot ignore the serious side-effects. In consumer products, like food and beverages, besides taste, different responses like colour, texture and the undesirable by-products have to be taken into account. It is therefore a challenging problem to extend the study of mixture experiments to the case of multiple responses. In this connection, Mandal and Pal [17] studied the problem of determining A-optimal designs for estimating the optimum proportions in a multi-response experiment.

It is also possible to have situations where the response depends not only on the mixing proportions, but also on the amount of the mixture. An example is the effect of a fertilizer on the yield of a crop, which depends not only on the composition of the fertilizer but also on the amount of fertilizer applied. To date, there are only a few studies on such mixture-amount models in the single response case. See, for example, Hilgers and Bauer [18], Heiligers and Hilgers [19], Zhang et al. [20], Mandal et al. [21], Pal and Mandal [22], Mandal and Pal [23]. In this paper, we study the problem of determining optimal designs for the estimation of optimum proportions and amount for each response in a multi-response mixture experiment, where each response is defined by the mixture-amount model that has been proposed by Pal and Mandal [22]. The paper is organized as follows. Section 2 describes the problem and its perspectives. Section 3 investigates the optimal designs using a linear optimality criterion, namely the trace criterion. Finally, a discussion on our findings is given in Section 4.

2. The problem and Its Perspective

Consider a mixture experiment with q components, whose proportions in the mixture are denoted by $x_1, x_2, ..., x_q$, and the amount of mixture used is A. The experimental domain is given by

$$\Xi = \{(A, x_1, x_2, ..., x_q) \mid A \in [A_L, A_U], A_L > 0, x_i \geq 0, i = 1, 2, \ldots, q, \sum_i x_i = 1\}. \tag{2.1}$$

The assumption $A_L > 0$ ensures that some amount of the mixture should necessarily be used in the experiment.

Let $\boldsymbol{Y}' = (Y^{(1)}, Y^{(2)}, \ldots, Y^{(p)})$ be the random response vector, where $Y^{(g)}$ denotes the g^{th} characteristic of the output, $g = 1, 2, \ldots, p$. We assume that $Y^{(g)}$ is dependent on the proportions $x_1, x_2, ..., x_q$ and the amount A, and its mean is a second degree polynomial in $\boldsymbol{x^*} = (A, x_1, x_2, ..., x_q)$:

$$E(Y^{(g)} \mid \boldsymbol{x^*}) = \varsigma_{x^*}^{(g)} = \alpha_{01}^{(g)} A + \alpha_{02}^{(g)} A^2 + A \sum_{i=1}^{q} \alpha_{0i}^{(g)} x_i + \sum_{i=1}^{q} \alpha_{ii}^{(g)} x_i^2 + \sum_{i<j=1}^{q} \alpha_{ij}^{(g)} x_i x_j \tag{2.2}$$

This model has been suggested by Pal and Mandal [22] in the single response case.

Using the constraint $\sum_i x_i = 1$, we can write (2.2) as

$$\varsigma_{x^*}^{(g)} = f'(\boldsymbol{x}) \boldsymbol{\beta}^{(g)} = \boldsymbol{x}^{*\prime} B^{(g)} \boldsymbol{x}^* \tag{2.3}$$

where

$$\boldsymbol{f}(\boldsymbol{x}) = (A^2, Ax_1, Ax_2, ..., Ax_q, x_1^2, x_2^2, ..., x_q^2, x_1x_2, x_1x_3, ..., x_{q-1}x_q)'$$

$$\boldsymbol{\beta}^{(g)} = (\beta_{00}, \beta_{01}, \beta_{02}, ..., \beta_{0q}, \beta_{11}, \beta_{22}, ..., \beta_{qq}, \beta_{12}, ..., \beta_{q-1q})'$$

$$B^{(g)} = ((b_{ij}^{(g)})),$$

$$b_{ij}^{(g)} = \beta_{ii}^{(g)}, \text{ if } i = j$$

$$= \frac{1}{2}\beta_{ij}^{(g)}, \text{ if } i < j.$$

Here $\beta_{ij}^{(g)}$ s being linear functions of $\alpha_{ij}^{(g)}$ s.

Here, $\boldsymbol{x}^*$ satisfies the constraint

$$\boldsymbol{c}'\boldsymbol{x}^* = 1, \tag{2.4}$$

where $\boldsymbol{c} = (0,1, 1, \ldots, 1)'$.

We assume that for each $g = 1, 2, \ldots, p,$ $\varsigma_{x^*}^{(g)}$ is concave and has a finite maximum in the interior of the experimental region (2.1). Then, subject to (2.4), (2.3) is maximized at

$$\boldsymbol{\gamma}^{(g)*} = \delta^{(g)^{-1}} B^{(g)^{-1}} \boldsymbol{c}, \tag{2.5}$$

where

$$\delta^{(g)} = \boldsymbol{c}' \; B^{(g)^{-1}} \boldsymbol{c}.$$

Let us write $\boldsymbol{\gamma}^{(g)*} = (A^{(g)}, \gamma_1^{(g)}, ..., \gamma_q^{(g)})$, where $A^{(g)}$ corresponds to the optimum amount and $(\gamma_1^{(g)}, \gamma_2^{(g)}, ..., \gamma_q^{(g)})$ the optimum mixing proportions for g-th response.

We are interested in estimating the non-linear functions $\boldsymbol{\gamma}^{(g)*}$, $g = 1, 2, \ldots, p$, 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.

We can write

$$E(\boldsymbol{Y}) = \Theta f(\boldsymbol{x}), \tag{2.6}$$

with

$$\Theta = (\boldsymbol{\beta}^{(1)}, \boldsymbol{\beta}^{(2)}, \ldots, \boldsymbol{\beta}^{(p)})'.$$

Let $\Sigma = ((\sigma_{gh}))$ denote the dispersion matrix of $\boldsymbol{\gamma}$.

For any arbitrary continuous design ξ in Ξ, the information matrix of ξ for $\boldsymbol{\beta} = (\boldsymbol{\beta}^{(1)\prime}, \boldsymbol{\beta}^{(2)\prime}, \ldots, \boldsymbol{\beta}^{(p)\prime})'$ is given by

$$I(\xi, \boldsymbol{\beta}) = \Sigma^{-1} \otimes M(\xi),$$

where $M(\xi) = \int_{\Xi} f(\boldsymbol{x}) f'(\boldsymbol{x}) d\xi(\boldsymbol{x})$.

For a given design ξ, we can estimate Θ by $\hat{\Theta}$, the least squares estimator of Θ, and hence $B^{(g)}$ by $\hat{B}^{(g)}$ and $\delta^{(g)}$ by $\hat{\delta}^{(g)}$. Then, an estimate of $\gamma^{(g)*}$ is given by

$$\hat{\gamma}^{(g)*} = \hat{\delta}^{(g)^{-1}} \hat{B}^{(g)^{-1}} \mathbf{1},\ g = 1, 2, \ldots, p, \tag{2.7}$$

where $\mathbf{1}$ is a unit vector. Let, $\gamma^* = (\gamma^{(1)*\prime}, \gamma^{(2)*\prime}, \ldots, \gamma^{(g)*\prime})'$. Then, under suitable regularity assumptions on error distribution, the standard ∂-method gives an adequate approximation of the dispersion matrix of $\hat{\gamma}^* = (\hat{\gamma}^{(1)*\prime}, \hat{\gamma}^{(g)*\prime}, \ldots, \hat{\gamma}^{(g)*\prime})'$ as

$$\begin{aligned} Disp(\hat{\gamma}^*) &= E[(\hat{\gamma}^* - \gamma^*)(\hat{\gamma}^* - \gamma^*)'] \\ &= Diag(T^{(g)}(\gamma^*), g = 1,2,\ldots, p)(\Sigma \otimes M^{-1}(\xi)) Diag(T^{(g)}(\gamma^*), g = 1,2,\ldots, p)' \end{aligned} \tag{2.8}$$

where

$$T^{(g)}(\gamma^*) = \left(\frac{\partial \gamma^{(g)*}}{\partial \beta_{00}^{(g)}}, \frac{\partial \gamma^{(g)*}}{\partial \beta_{01}^{(g)}}, \frac{\partial \gamma^{(g)*}}{\partial \beta_{02}^{(g)}}, \frac{\partial \gamma^{(g)*}}{\partial \beta_{11}^{(g)}}, \ldots, \frac{\partial \gamma^{(g)*}}{\partial \beta_{qq}^{(g)}}, \frac{\partial \gamma^{(g)*}}{\partial \beta_{12}^{(g)}}, \ldots, \frac{\partial \gamma^{(g)*}}{\partial \beta_{q-1,q}^{(g)}} \right) \tag{2.9}$$

$$= \begin{bmatrix} 0 & -(q-1)A^{(g)}/2 & \cdots & A^{(g)}/2 & -(q-1)\gamma_1^{(g)} & \cdots & \gamma_q^{(g)} & \frac{1}{2}\gamma_1^{(g)} - \frac{q-1}{2}\gamma_2^{(g)} & \cdots & \frac{1}{2}(\gamma_{q-1}^{(g)} + \gamma_q^{(g)}) \\ 0 & A^{(g)}/2 & \cdots & A^{(g)}/2 & \gamma_1^{(g)} & \cdots & \gamma_q^{(g)} & \frac{1}{2}\gamma_2 - \frac{q-1}{2}\gamma_1^{(g)} & \cdots & \frac{1}{2}(\gamma_{q-1}^{(g)} + \gamma_q^{(g)}) \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ 0 & A^{(g)}/2 & \cdots & A^{(g)}/2 & \gamma_1^{(g)} & \cdots & \gamma_q^{(g)} & \frac{1}{2}(\gamma_1^{(g)} + \gamma_2^{(g)}) & \cdots & \frac{1}{2}\gamma_{q-1}^{(g)} - \frac{q-1}{2}\gamma_q^{(g)} \\ 0 & A^{(g)}/2 & \cdots & -(q-1)A^{(g)}/2 & \gamma_1^{(g)} & \cdots & -(q-1)\gamma_q^{(g)} & \frac{1}{2}(\gamma_1^{(g)} + \gamma_2^{(g)}) & \cdots & \frac{1}{2}\gamma_q^{(g)} - \frac{q-1}{2}\gamma_{q-1}^{(g)} \\ -qA^{(g)} & -q\gamma_1^{(g)}/2 & \cdots & -q\gamma_q^{(g)}/2 & 0 & \cdots & 0 & 0 & \cdots & 0 \end{bmatrix}$$

(2.10)

Here $A^{(g)}$ denotes the optimum amount and corresponding to the g^{th} response, $g = 1, 2, ..., p$.

We restrict our study to the class M of positive definite matrices $M(\xi)$. Since $Disp(\hat{\boldsymbol{\gamma}}^*)$ is singular, the trace criterion would be an appropriate criterion for comparing different designs:

$$\phi(\boldsymbol{\gamma}^*, M(\xi)) = tr(Disp(\hat{\boldsymbol{\gamma}}^*)). \tag{2.11}$$

However, $\boldsymbol{\gamma}^*$ being a non-linear function of the model parameters, (2.11) will involve these unknown parameters. A way out would be to adopt the pseudo-Bayesian approach of Pal and Mandal [13].

We assume that for each g, $\boldsymbol{\gamma}^{(g)*}$ is random with

$$\mathcal{E}(\gamma_i^{(g)2}) = v^{(g)}, \; i = 1, 2, ..., q; \quad \mathcal{E}(\gamma_i^{(g)}\gamma_j^{(g)}) = w^{(g)}, \; i \neq j = 1, 2, \ldots, q; \; v^{(g)} > 0,$$
$$w^{(g)} > 0, \; w^{(g)} \leq v^{(g)}, \tag{2.12}$$

and

$$\mathcal{E}(A^{(g)2}) = a^{(g)}, \; \mathcal{E}(A^{(g)}) = c^{(g)}, \; \mathcal{E}(A^{(g)}\gamma_i^{(g)}) = b^{(g)}, \; i = 1, 2, .., q, \; g = 1, 2, ..., p. \tag{2.13}$$

Since for each g, nothing is known about the relative influence of the different components $(A^{(g)}, \gamma_1^{(g)}, i = 1, 2, \ldots, q)$ of $\boldsymbol{\gamma}^{(g)*}$, there is no basis for assuming apriori moments to be unequal for the components. We therefore assume all first order and second order moments (pure and mixed) to be invariant with respect to the different components. In view of (2.4), the a-priori moments satisfy the following relations:

$1/q^2 < v^{(g)} < 1/q$, $q\, v^{(g)} + q(q\text{-}1)w^{(g)} = 1$, $qb^{(g)} = c^{(g)}$ and $a^{(g)} > c^{(g)\,2}$, $g = 1, 2,..., p$.

Thus, we consider the pseudo-Bayesian trace criterion given by

$$\psi(\xi) = \mathcal{E}\,[\,\phi(\gamma^*, M(\xi))\,],$$

where $\mathcal{E}$ is the expectation taken with respect to the prior distribution.

3. Pseudo-Bayesian Trace-Optimal Design

The criterion function can be written as

$$\psi(\xi) = \mathcal{E}\,[\,\phi(\gamma^*, M(\xi))\,]$$

$$= tr[(\Sigma \otimes M^{-1}(\xi))\,\mathcal{E}$$

$$\{Diag(T^{(g)}(\gamma^*), g = 1,2,..., p)' Diag(T^{(g)}(\gamma^*), g = 1,2,..., p)\}]$$

$$= tr[(\Sigma \otimes M^{-1}(\xi))\ \mathcal{E}\{Diag(T^{(g)}(\gamma^*)'T^{(g)}(\gamma^*)), g = 1,2,..., p)\}]$$

$$= \sum_g \sigma_{gg}\, tr[M^{-1}(\xi)\ \mathcal{E}\ \{T^{(g)}(\gamma^*)'T^{(g)}(\gamma^*)\}]$$

$$= tr[M^{-1}(\xi) \sum_g \sigma_{gg}\ \ \mathcal{E}\{T^{(g)}(\gamma^*)'T^{(g)}(\gamma^*)\}]\ , \qquad (3.1)$$

where

$$\mathcal{E}\,(T^{(g)}(\gamma^*)'T^{(g)}(\gamma^*))$$

$$= \begin{bmatrix} q^2 a^{(g)} & \frac{1}{2} q^2 b^{(g)} \underline{1}_q' & \underline{0}' & \underline{0}' \\ \frac{1}{2} q^2 b^{(g)} \underline{1}_q & a_1^{(g)} I_q + b_1^{(g)} \underline{1}_q \underline{1}_q' & a_2^{(g)} I_q + b_2^{(g)} \underline{1}_q \underline{1}'_q & D_1^{(g)} \\ \underline{0} & a_2^{(g)} I_q + b_2^{(g)} \underline{1}_q \underline{1}'_q & a_3^{(g)} I_q + b_3^{(g)} \underline{1}_q \underline{1}'_q & D_2^{(g)} \\ \underline{0} & D_1^{(g)\prime} & D_2^{(g)\prime} & D_3^{(g)} \end{bmatrix},$$

$$a_1^{(g)} = q(q-1)\frac{a^{(g)}}{4} + q^2\frac{v^{(g)}}{4} - b_1^{(g)},\ b_1^{(g)} = -\frac{a^{(g)}}{4}q + q^2\frac{w^{(g)}}{4},$$

$$a_2^{(g)} = \frac{q(q-1)}{2}b^{(g)} + \frac{b^{(g)}}{2}q, \quad b_2^{(g)} = -q\frac{b^{(g)}}{2},$$

$$a_3^{(g)} = q(q-1)v^{(g)} + qw^{(g)},\ b_3^{(g)} = \text{-}qw^{(g)}\ ,$$

$D_1^{(g)}, D_2^{(g)}$ and $D_3^{(g)}$ are $q \times C(q,2)$, $q \times C(q,2)$ and $C(q,2) \times C(q,2)$ matrices, respectively, given by

$$D_1^{(g)} = \frac{b^{(g)}}{4}\begin{bmatrix} d_1^{(g)} & d_1^{(g)} & d_1^{(g)} & \dots & d_1^{(g)} & d_2^{(g)} & d_2^{(g)} & \dots & d_2^{(g)} & d_2^{(g)} & \dots & d_2^{(g)} & d_2^{(g)} \\ d_1^{(g)} & d_1^{(g)} & d_1^{(g)} & \dots & d_2^{(g)} & d_1^{(g)} & d_1^{(g)} & \dots & d_1^{(g)} & d_2^{(g)} & \dots & d_2^{(g)} & d_2^{(g)} \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ d_2^{(g)} & d_2^{(g)} & d_2^{(g)} & \dots & d_2^{(g)} & d_2^{(g)} & d_2^{(g)} & \dots & d_1^{(g)} & d_2^{(g)} & \dots & d_2^{(g)} & d_1^{(g)} \end{bmatrix},$$

$$D_2^{(g)} = \frac{1}{2}\begin{bmatrix} d_3^{(g)} & d_3^{(g)} & d_3^{(g)} & \dots & d_3^{(g)} & d_4^{(g)} & d_4^{(g)} & \dots & d_4^{(g)} & d_4^{(g)} & \dots & d_4^{(g)} & d_4^{(g)} \\ d_3^{(g)} & d_4^{(g)} & d_4^{(g)} & \dots & d_4^{(g)} & d_3^{(g)} & d_3^{(g)} & \dots & d_3^{(g)} & d_4^{(g)} & \dots & d_4^{(g)} & d_4^{(g)} \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ d_4^{(g)} & d_4^{(g)} & d_4^{(g)} & \dots & d_3^{(g)} & d_4^{(g)} & d_4^{(g)} & \dots & d_3^{(g)} & d_4^{(g)} & \dots & d_4^{(g)} & d_3^{(g)} \end{bmatrix},$$

$$D_3^{(g)} = \frac{1}{4}\begin{bmatrix} d_5^{(g)} & d_6^{(g)} & \dots & d_6^{(g)} & d_6^{(g)} & d_6^{(g)} & \dots & d_6^{(g)} & d_7^{(g)} & \dots & d_7^{(g)} & \dots & d_7^{(g)} & d_7^{(g)} \\ d_6^{(g)} & d_5^{(g)} & \dots & d_6^{(g)} & d_6^{(g)} & d_7^{(g)} & \dots & d_7^{(g)} & d_6^{(g)} & \dots & d_6^{(g)} & \dots & d_7^{(g)} & d_7^{(g)} \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ d_7^{(g)} & d_7^{(g)} & \dots & d_6^{(g)} & d_7^{(g)} & d_7^{(g)} & \dots & d_6^{(g)} & d_7^{(g)} & \dots & d_6^{(g)} & \dots & d_6^{(g)} & d_5^{(g)} \end{bmatrix},$$

$$d_1^{(g)} = q(q-2),\ d_2^{(g)} = -2q,$$

$$d_3^{(g)} = q\{-v^{(g)} + (q-1)w^{(g)}\},\ d_4^{(g)} = -2qw^{(g)}$$

$$d_5^{(g)} = 2q(q-1)v^{(g)} - 2qw^{(g)},\ \ d_6^{(g)} = -q(v^{(g)} - \overline{q-3}w^{(g)}),\ d_7^{(g)} = -4qw^{(g)}\ .$$

It may be noted that $\psi\ (\xi)$ is a convex function of $M(\xi)$ in M and (3.1) is invariant with respect to the mixing proportions. Hence, the optimum design will also be invariant with respect to x_i s. Therefore, for a given value of A, we can confine our search within the class of weighted centroid designs [24, 25]. Further, for x_i s given, since the model (2.6) is quadratic in A, the optimum design is likely to admit three distinct values of A, two at the two extremes and one in between, with positive weights. We may, therefore, initially confine our search for an optimal design within the sub-class D_q of designs having support points and weights as given in Table 3.1.

Table 3.1. Support points and corresponding weights of a typical design in D_q.

x_1	x_2	…	x_{q-1}	x_q	weight	A	weight
1	0	…	0	0	p_1		
0	1	…	0	0	p_1		
…	…	…	…	…	…		
0	0	…	0	1	p_1	-1	w_{-1}
½	½	…	0	0	p_2		
½	0	…	0	0	p_2		
…	…	…	…	…	…		
0	0	…	½	½	p_2		
1	0	…	0	0	p_1'		
0	1	…	0	0	p_1'		
…	…	…	…	…	…		
0	0	…	0	1	p_1'	a_0	w_0
½	½	…	0	0	p_2'		
½	0	…	0	0	p_2'		
…	…	…	…	…	…		
0	0	…	½	½	p_2'		
1	0	…	0	0	p_1''		
0	1	…	0	0	p_1''		
…	…	…	…	…	…		
0	0	…	0	1	p_1''	1	w_1
½	½	…	0	0	p_2''		
½	0	…	0	0	p_2''		
…	…	…	…	…	…		
0	0	…	½	½	p_2''		

Here, $0 \le p_i, p_i', p_i'' \le 1$, $i = 1,2$, $C(q, 1)p_1 + C(q, 2)p_2 = 1$, $C(q, 1)p_1' + C(q, 2)p_2' = 1$, $C(q, 1)\, p_1'' + C(q, 2)\, p_2'' = 1$, $a_0 \in (-1, 1)$, $w_j \ge 0$, $j = -1, 0, 1$, $w_{-1} + w_0 + w_1 = 1$, and $C(q,k) = \binom{q}{k}$. w_{-1}, w_0 and w_1 denote the weights attached to $A = -1$, a_0, 1 respectively, while the sixth column gives the weights for different $(x_1, x_2, ..., x_q)$ combinations when A is given.

For any design $\xi \in \mathrm{D}_q$, the moment matrix can be written as

$$M(\xi) = \begin{bmatrix} M_{11} & M_{12} & M_{13} \\ M_{12}' & M_{22} & M_{23} \\ M_{13}' & M_{23}' & M_{33} \end{bmatrix},$$

where

$$M_{11} = \begin{bmatrix} w_1 + w_{-1} + a_0^4 w_0 & a_{11}\underline{\mathbf{1}}_q' \\ & b_{11} I_q + c_{11}\underline{\mathbf{1}}_q \underline{\mathbf{1}}_q' \end{bmatrix}, \; M_{12} = \begin{bmatrix} a_{12}\underline{\mathbf{1}}_q' \\ b_{12} I_q + c_{12}\underline{\mathbf{1}}_q \underline{\mathbf{1}}_q' \end{bmatrix}$$

$$M_{13} = \begin{bmatrix} a_{13}\mathbf{1}_{C(q,2)}' \\ c_{13} M_0 \end{bmatrix}, \; M_{22} = b_{22} I_q + c_{22}\mathbf{1}_q \mathbf{1}_q',$$

$$M_{23} = c_{23} M_0, \; M_{33} = b_{33} I_{C(q,2)},$$

$$a_{11} = \{p_1 + \frac{q-1}{2} p_2\} w_1 - \{p_1'' + \frac{q-1}{2} p_2''\} w_{-1} + a_0^3 \{p_1' + \frac{q-1}{2} p_2'\} w_0$$

$$b_{11} = \{p_1 + \frac{q-2}{2} p_2\} w_1 + \{p_1'' + \frac{q-2}{2} p_2''\} w_{-1} + a_0^2 \{p_1' + \frac{q-2}{2} p_2'\} w_0$$

$$c_{11} = \frac{p_2}{4} w_1 + \frac{p_2''}{4} w_{-1} + a_0^2 \frac{p_2'}{4} w_0$$

$$a_{12} = \{p_1 + \frac{q-1}{4} p_2\} w_1 + \{p_1'' + \frac{q-1}{4} p_2''\} w_{-1} + a_0^2 \{p_1' + \frac{q-1}{4} p_2'\} w_0$$

$$b_{12} = \{p_1 + \frac{q-2}{8} p_2\} w_1 - \{p_1'' + \frac{q-2}{2} p_2''\} w_{-1} + a_0 \{p_1' + \frac{q-2}{8} p_2'\} w_0$$

$$c_{12} = \frac{p_2}{8} w_1 - \frac{p_2''}{8} w_{-1} + a_0 \frac{p_2'}{8} w_0$$

$$a_{13} = 2\{\frac{p_2}{8} w_1 - \frac{p_2''}{8} w_{-1} + a_0^2 \frac{p_2'}{8} w_0\}$$

$$c_{13} = \frac{p_2}{8} w_1 - \frac{p_2''}{8} w_{-1} + a_0 \frac{p_2'}{8} w_0$$

$$b_{22} = \{p_1 + \frac{q-2}{16} p_2\} w_1 + \{p_1'' + \frac{q-2}{16} p_2''\} w_{-1} + \{p_1' + \frac{q-2}{16} p_2'\} w_0$$

$$c_{22} = \frac{p_2}{16} w_1 + \frac{p_2''}{16} w_{-1} + \frac{p_2'}{16} w_0 = c_{23} = b_{33},$$

M_0 is a $q \times C(q, 2)$ matrix in which the first q-1 elements in the first row are 1 and the remaining are 0, and the following q-1 rows are permutations of the first row.

Since for quadratic regression in [-1, 1], the optimum support points of D-, A- and E- optimality criteria are at -1, 0 and 1, with equal weights at the extreme points, to start with, we take $a_0 = 0$, and $w_{-1} = w_1$, $p_i = p_i''$, $i = 1, 2$. Let, $\mathcal{D}_q^0 \subset D_q$ define the corresponding subclass of designs.

Then, for any design $\xi \in \mathcal{D}_q^0$, we have

$$M(\xi) = \begin{array}{c} \begin{array}{cccccccccccccc} A^2 & Ax_1 & Ax_2 & \ldots & \ldots & Ax_q & x_1^2 & x_2^2 & \ldots & \ldots & x_q^2 & x_1x_2 & x_1x_3 & \ldots x_{q-1}x_q \end{array} \\ \hline \begin{bmatrix} 2w_1 & \mathbf{0}_q' & 2w_1(p_1 + \frac{q\text{-}1}{4} p_2)\mathbf{1}_q' & 2w_1 \frac{p_2}{4} \mathbf{1}_{C(q,2)}' \\ & 2w_1\{(p_1 + \frac{q\text{-}2}{4} p_2) I_q + \frac{p_2}{4} \mathbf{1}_q \mathbf{1}_q'\} & 0 & 0 \\ & & r_1 I_q + r_2 \mathbf{1}_q \mathbf{1}_q' & r_2 M_0 \\ & & & r_2 I_{C(q,2)} \end{bmatrix} \end{array},$$

where

$$r_1 = 2w_1(p_1 + \frac{q-2}{16} p_2) + (1 - 2w_1)(p_1' + \frac{q-2}{16} p_2')$$

$$r_2 = 2w_1 \frac{1}{16} p_2 + (1 - 2w_1) \frac{1}{16} p_2'.$$

Therefore, $M^{-1}(\xi)=\begin{bmatrix} M_{11}^{*} & M_{12}^{*} \\ M_{21}^{*} & M_{22}^{*} \end{bmatrix}$, where

$$M_{11}^{*}=\frac{1}{2w_1}\begin{bmatrix} \frac{1}{1-2w_1k_1}1 & \mathbf{0}' \\ \mathbf{0} & k_2I_q+k_3\mathbf{1}_q\mathbf{1}_q' \end{bmatrix},$$

$$M_{12}^{*}=-\frac{1}{(1-2w_1k_1)}\begin{bmatrix} t\mathbf{1}_q' & s\mathbf{1}_{C(q,2)}' \\ 0 & 0 \end{bmatrix},$$

$$M_{22}^{*}=\begin{bmatrix} \frac{1}{r}I_q & -\frac{1}{r}M_0 \\ -\frac{1}{r}M_0 & \frac{1}{r_2}I_{C(q,2)}+\frac{1}{r}M_0'M_0 \end{bmatrix}+\frac{2w_1}{1-2w_1k_1}\begin{bmatrix} t^2\mathbf{1}_q\mathbf{1}_q' & ts\mathbf{1}_q\mathbf{1}_{C(q,2)}' \\ st\mathbf{1}_{C(q,2)}\mathbf{1}_q' & s^2\mathbf{1}_{C(q,2)}\mathbf{1}_{C(q,2)}' \end{bmatrix},$$

$$k_1=\frac{q}{r}p_1^2+\frac{C(q,2)}{r_2}\frac{p_2^2}{16},\quad k_2=\frac{1}{p_1+\frac{q\text{-}2}{4}p_2},k_3=-\frac{p_2/4}{(p_1+\frac{q-2}{4}p_2)(p_1+\frac{q-1}{2}p_2)},$$

$$t=\frac{p_1}{r},\quad s=-\frac{2}{r}p_1+\frac{p_2}{4r_2},\quad r=2w_1p_1+(1-2w_1)p_1'.$$

We thus get

$$\begin{aligned}\psi(\xi)=\sum_g\sigma_{gg}\{q^2\frac{a^{(g)}}{2w_1(1-2w_1k_1)}+q\frac{a_1^{(g)}(k_2+k_3)+b_1^{(g)}(k_2+qk_3)}{2w_1}\\
+q\frac{2w_1^{(g)}}{1-2w_1^{(g)}k_1}[t\{t(a_3^{(g)}+qb_3^{(g)})+(q-1)s(d_3^{(g)}+\frac{q-2}{2}d_4^{(g)})\}\\
+\frac{q-1}{8}s^2\{d_5^{(g)}+2(q-2)d_6^{(g)}+\frac{(q-2)(q-3)}{2}d_7^{(g)}\}]+q\frac{a_3^{(g)}+b_3^{(g)}-(q-1)d_3^{(g)}}{r}\\
+\frac{q(q\text{-}1)d_5^{(g)}}{8r_2}+\frac{q(q-1)}{8r}[2\{d_5^{(g)}+(q-2)d_6^{(g)}\}]\}\end{aligned}$$

(3.2)

The optimal values of p_1, p_1' and w_1 are obtained by minimizing $\psi(\xi)$. The optimality of the design thus obtained within the whole class of competing designs can be checked using the multi-variate version of the Equivalence Theorem, given in Pal and Mandal [22] for the single response mixture amount model:

Equivalence Theorem 3.1: A necessary and sufficient condition for a design ξ^* to be trace-optimal in a p-variate regression model is that

$$f(\boldsymbol{x})'M^{-1}(\xi^*)[\sum_{g=1}^{p}\sigma_{gg}\,\mathcal{E}(T^{(g)\prime}T^{(g)})]M^{-1}(\xi^*)\,f(\boldsymbol{x}) \le \phi(\xi^*) \qquad (3.3)$$

for all $\boldsymbol{x} \in \Xi$. Equality in (3.3) holds at all the support points of ξ^*.

As the algebraic derivations are rather involved, the condition (3.3) may be checked by numerical computation. We give below in Table 3.2 the optimum designs for q-component model, $2 \le q \le 4$, which have been numerically examined to satisfy condition (3.3), for various combinations of $(\sigma_{gg}, g = 1,2,..., p)$ and the apriori moments $((a^{(g)}, v^{(g)}, w^{(g)}), g = 1, 2,\ldots, p)$. The designs for q ≥ 5 may be similarly obtained and their optimality or otherwise checked numerically using Theorem 3.1.

Table 3.2. Optimum designs for $q = 2, 3, 4$; $p = 2, 3, 4$ and some combinations of $(\sigma_{gg}, a^{(g)}, v^{(g)}, w^{(g)})$ s.

q	p	$(a^{(1)},\ldots,a^{(p)})$	$(\sigma_{11},\cdots\sigma_{pp})$	$(v^{(1)},\ldots,v^{(p)})$	p_1	p_1'	w_1	w_0	Trace
2	2	(0.2, 0.4)	(1, 3)	(0.30, 0.40)	0.29915	0.27692	0.31869	0.36261	122.780
	2	(0.2, 0.6)	(1,10)	(0.27, 0.45)	0.29609	0.27151	0.31246	0.37507	482.261
	3	(0.2, 0.6, 0.8)	(1, 3, 7)	(0.27, 0.32, 0.40)	0.31207	0.28381	0.30335	0.39329	407.639
	3	(0.1, 0.5, 0.7)	(1, 6, 10)	(0.30, 0.37, 0.45)	0.29988	0.27424	0.37908	0.38043	706.915
	4	(0.1, 0.3, 0.5, 0.7)	(1, 4, 8, 10)	(0.28, 0.32, 0.4, 0.48)	0.29785	0.27321	0.31226	0.37548	934.674
	4	(0.1, 0.5, 0.7, 0.9)	(1, 6, 10, 14)	(0.3, 0.37, 0.45, 0.26)	0.32015	0.29093	0.30062	0.39876	1,075.68
3	2	(0.2, 0.4)	(1, 3)	(0.15, 0.25)	0.12969	0.10904	0.35186	0.29628	973.012
	2	(0.2, 0.6)	(1,10)	(0.24, 0.32)	0.13363	0.11377	0.35000	0.30000	4,301.49
	3	(0.2, 0.6, 0.8)	(1, 3, 7)	(0.14, 0.24, 0.32)	0.13552	0.11149	0.34406	0.31188	3,902.46
	3	(0.1, 0.5, 0.7)	(1, 6, 10)	(0.12, 0.20, 0.33)	0.13423	0.11132	0.34596	0.30807	5,647.98
	4	(0.1, 0.3, 0.5, 0.7)	(1, 4, 8, 10)	(0.12, 0.18, 0.22, 0.30)	0.13339	0.10947	0.34554	0.30892	6,562.84
	4	(0.1, 0.5, 0.7, 0.9)	(1, 6, 10, 14)	(0.15, 0.20, 0.25, 0.32)	0.13624	0.11049	0.34218	0.31564	10,489.7
4	2	(0.6, 0.2)	(1, 3)	(0.08, 0.0.18)	0.07387	0.05906	0.37747	0.24505	4,118.26
	2	(0.2, 0.4)	(1,10)	(0.10, 0.15)	0.07456	0.05640	0.37262	0.25476	10,645.4
	3	(0.2, 0.6, 0.8)	(1, 3, 7)	(0..07, 0..14, 0..24)	0.07877	0.05887	0.36902	0.26196	16,029.4
	3	(0.1, 0.5, 0.7)	(1, 6, 10)	(0.08, 0.18, 0.22)	0.07880	0.05888	0.36899	0.26202	24,836.1
	4	(0.1, 0.3, 0.5, 0.7)	(1, 4, 8, 10)	(0.07, 0.12, 0.18, 0.22)	0.07724	0.05899	0.37132	0.25735	29,931.0
	4	(0.1, 0.5, 0.7, 0.9)	(1, 6, 10, 14)	(0.09, 0.15, 0.20, 0.24)	0.07913	0.05914	0.36879	0.26242	47,361.1

4. Conclusion

In this paper, we have investigated the problem of finding the optimum design for estimation of optimum mixing proportions and the optimum mixture amount in a multi-response mixture experiment, when each response function is quadratic, concave. The trace-optimal design has been obtained numerically for the number of components q = 2, 3, 4, and is seen to belong to the subclass D_q of designs given by Table 3.1. The weights assigned to the support points of the design are dependent on the *apriori* moments of the optimum proportions and the optimum amounts. It may be conjectured that for higher values of q, the optimal design will also belong to the subclass D_q.

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