Special Proceedings: ISBN #: 978-81-950383-5-0
26th Annual Conference, 26-28 February 2024; 97-110

Optimum Mixture Designs in Constrained Experimental Regions - An Informative Review

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Received: 22 May 2024; Revised: 30 June 2024; Accepted: 02 July 2024

Abstract

In a mixture experiment, the response depends on the proportions of the mixing components. Canonical models of different degrees have been suggested by Scheffé (1958) to represent the mean response in terms of the mixing proportions, and optimum designs for estimation of the parameters of the models have been investigated by several authors. In most cases, the optimum design includes the vertex points of the simplex as support points of the design, which are not mixture combinations in the true non-trivial sense, and therefore are not acceptable to the practitioners. Further, in some situations, due to physical or economic limitations, the experimental region forms only a part of the simplex that does not cover the extreme points. The present paper gives a review of the available literature on optimum mixture experiments in regular subspaces of the simplex.

Key words: Mixture experiments; Restricted experimental region; Optimum designs.

AMS Subject Classifications: 62K99, 62J05

1. Introduction

A systematic study of optimum regression designs began with the pathbreaking work of Kiefer and Wolfowitz (1959). Soon after, various authors started to investigate optimality criteria for designs to estimate the model parameters (*cf.* Elfving, 1959; Karlin and Studden, 1966; Fedorov, 1971; Pukelsheim, 1993; Draper and Pukelsheim, 1996; Liski *et al.* 1998; Li *et al.*, 2005). A mixture experiment is a special case of a regression experiment, where the mean response is dependent on the mixing proportions of the ingredients in the mixture, rather than on their actual amounts. Thus, for a mixture experiment with q ingredients, the experimental region is defined by

$$\Xi = \left\{ (x_1, x_2, \ \ldots, \ x_q)^T : \ x_i \geq 0, \ i = 1\,(1)\,q, \ \sum_{i=1}^{q} x_i = 1 \right\}, \tag{1}$$

where $(x_1, x_2, \ \ldots, \ x_q)$ denote the mixing proportions. Graphically, (1) is defined by a simplex with vertices $(1, 0, \ldots, 0), (0, 1, \ldots, 0), \ldots, (0, 0, \ldots, 1)$.

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There are high applications of mixture methodologies in different research areas, like (a) agricultural experiments, such as (i) intercropping (Dhekale *et al.*, 2003), (ii) split of total fertilizer application at different growth-stages of plants (Batra *et al.*, 1999), (iii) blend of waste water/saline water/marginal quality water for effective irrigation (Kan and Rapaport-Rom, 2012), (b) horticultural experiments, such as preparation of ready-to-serve beverages (Deka *et al.*, 2001), (c) animal nutritional experiments, such as feeding trials with several alternatives (Osborne and Mendel, 1921), (d) gasoline blending (Snee, 1981), (e) experiments with chemical pesticides (Deneer, 2000), and so on.

Mixture models, of the form $\eta_{\boldsymbol{x}} = \boldsymbol{f'(x)\beta}$, were first introduced by Scheffé (1958), who defined canonical models of degrees one to three to express the mean response in terms of the mixing proportions as follows:

Linear (homogeneous): $\eta_{\boldsymbol{x}} = \sum_i \beta_i x_i$

Quadratic: $\eta_{\boldsymbol{x}} = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j$

Full cubic: $\eta_{\boldsymbol{x}} = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j + \sum_{i<j<k} \beta_{ijk} x_i x_j x_k + \sum_{i<j} \delta_{ij} (x_i - x_j)$

Special cubic: $\eta_{\boldsymbol{x}} = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j + \sum_{i<j<k} \beta_{ijk} x_i x_j x_k$.

Scheffé (1958, 1963) also proposed the simplex lattice design and the simplex centroid design as suitable for parameter estimation in his proposed models. Later, other models, like the log-contrast model, Darroch-Waller quadratic mixture model, linear mixture models with synergism, were introduced.

Optimal designs for estimation of model parameters in various mixture models have been investigated by many researchers. Noteworthy are the studies by Kiefer (1961), Farrel *et al.* (1967), Atwood (1969), Galil and Kiefer (1977), Liu and Neudecker (1995), to name a few. Generally, the designs suggested for estimation and analysis in mixture experiments include the vertex points of the simplex, and such designs also turn out to be optimal designs. However, practitioners find such suggestions rather absurd and illogical as vertices of the simplex are not mixtures in the true sense, and they prefer to perform experiments excluding these points. Further, often due to physical or economic limitations, or interest of the experimenter, experiments may be confined to a sub-region of the whole experimental space. For example, when interest lies on the relationship among the ingredients, factorial arrangements can be used to analyze the response to ratios of ingredients (Kentworthy, 1963). Here only complete mixtures must be considered, that is, only mixtures where the proportion of each component is greater than zero. In agricultural/horticultural experiments, there are instances of usage of mixture experiments, and a growing interest in use of restricted subspaces of the simplex (Batra *et al.* 1999; Deka *et al.* 2001; Dhekale *et al.* 2003). Suggestion for the experimental region as a subspace of the simplex that does not include the vertex points are available in (Cornell, 2002). Though much research has been conducted to find appropriate designs for mixture experiments with restricted space, not much studies are available where the optimal design has been investigated.

This paper takes the readers on a journey through optimum designs when the experimental region is defined by a regular subspace of the simplex, such as an ellipsoid, a simplex within the simplex or a cuboid.

2. Restricted regions

The most common form of restricted region arises when one or more of the proportions of ingredients in the mixture are subjected to lower and/or upper bounds. This is very common in pharmaceutical experiments, horticulture experiments, agricultural experiments, gasoline blending, etc. The experimental region in such cases form a subset of the simplex. For example, if we have a 4-component mixture with the mixing proportions x_1, x_2, x_3 and x_4 having bounds $0.4 \leq x_1 \leq 0.6$, $0.1 \leq x_2, x_3 \leq 0.5$, $0.03 \leq x_4 \leq 0.08$, the experimental region is given by the bounded region within the simplex in Figure 1.

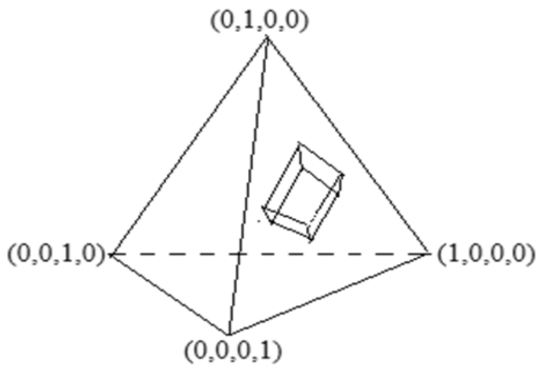


Figure 1: Experimental region within the simplex

In view of the bounds on the mixing proportions, the vertices of the simplex are excluded in the experimental region.

An interesting investigation carried out with such lower or upper bounds on mixing proportions, or on linear combinations of them, is due to Martin *et al.* (1999), who argued that theory cannot usually be used to obtain a good design. They discussed the algorithmic methods to obtain optimal designs, mainly using the D-optimality criterion, and sometimes the V – optimality criterion, and compared the algorithms using several published 3-component mixture examples. Their study was restricted to optimum designs for parameter estimation in Scheffé's canonical models. Later, Mandal *et al.* (2008) attempted to find the optimum design for estimation of the optimum mixing proportions in 2- and 3-component mixtures using Scheffé's quadratic mixture model, where one of the components is restricted by an upper bound less than unity. They used the pseudo-Bayesian approach due to Pal and Mandal (2006), and obtained the A-optimal design in the case of 2-component mixture. However, in the case of 3-component mixture, they could suggest an optimum design within six-point designs, but not within all competing designs. This instigated them to search further, and they came up with a seven-point design which was very close to the other design in terms of the criterion function. So, their suggestion was to start with any one of these designs, and use a standard numerical algorithm to reach the optimum design.

Other types of restricted experimental regions may be as given in Figure 2, As is noted, these regions have regular shapes, which are easy to study analytically, rather than very irregular regions within the simplex.

The restricted regions in Figure 2 also do not include the vertex points of the simplex.

An ellipsoidal experimental region often appears in pharmaceutical and engineering

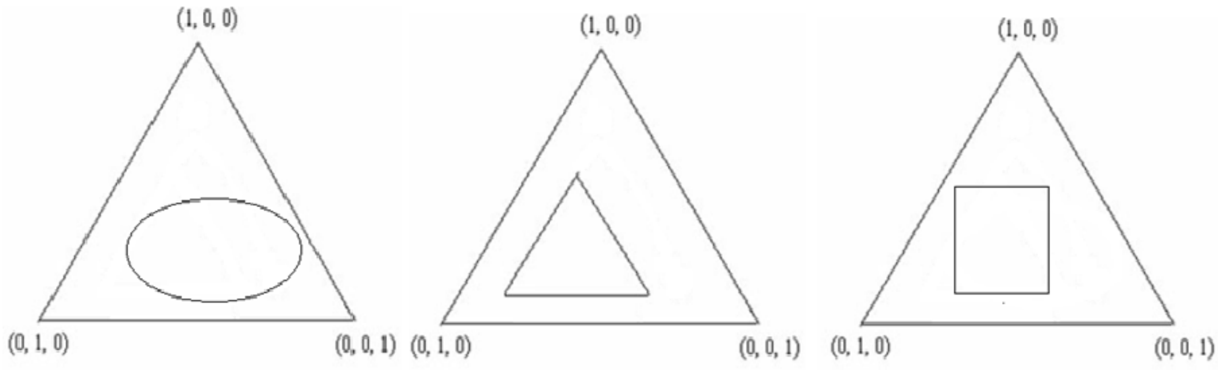


Figure 2: Some regular subspaces within the simplex forming the experimental regions

experiments. For example, Rais *et al.* (2004) used an ellipsoidal subregion of the space of mixture components for the optimization of a fluoroanhydrite-based self-levelling floor composition. A simplex within a simplex arises as an experimental region when the mixing proportions lie within a fixed range in (0,1). An example of this can be found in pharmaceutical experiment with oral tablets, where 2 or 3 polymers may be used with proportions having the same fixed non-zero bounds. Again, bounds on the mixing proportions may lead to a rectangular cuboid experimental region under certain conditions, as shown by Crosier (1990).

Restricted experimental region, ignoring the vertex points of the simplex, have been studies in mixture experiments to prescribe designs for parameter estimation (*cf.* Cornell, 2002). However, few authors attempted to find the optimum designs in such cases. Sections 3 - 5 review optimal designs for parameter estimation in Scheffé's first and second order models under regular experimental regions as indicated in Figure 2.

3. Ellipsoidal experimental region in the simplex

Mandal *et al.* (2015) were perhaps the first to attempt to find optimal design in an ellipsoidal region. For a q-component mixture experiment, they defined the constrained experimental region as

$$\Xi_0 = \left\{ \boldsymbol{x} = (x_1, x_2, \ldots, x_q)^T : x_i \geq 0, 1 \leq i \leq q, \sum_{i=1}^{q} x_i = 1, (\boldsymbol{x} - \boldsymbol{x}_0)^T H^{-2} (\boldsymbol{x} - \boldsymbol{x}_0) \leq 1 \right\}$$

where $\boldsymbol{x_0} = (1/q, 1/q, \ldots, 1/q)^T$ is the centroid of the simplex, and H is a non-singular diagonal matrix given by $H = diag\,(h_{11}, h_{22}, \ldots, h_{qq})$. The experimental region can be made to suit a specific situation by varying the h_{ii}'s.

The authors considered the case where $H \propto I_q$, an identity matrix. Then, for the transformation $\boldsymbol{x} \to \boldsymbol{z} = H^{-1}(\boldsymbol{x} - \boldsymbol{x_0})$, the domain of $\boldsymbol{z}$ comes out as $\{\boldsymbol{z} = (z_1, z_2, ..., z_n)^T : \boldsymbol{z}^T\boldsymbol{z} \leq 1, \boldsymbol{z}^T\boldsymbol{1}_q = 0\}$. A further transformation, *viz.* $\begin{bmatrix} u \\ \boldsymbol{v}^{(q-1)\times 1} \end{bmatrix} = Q\boldsymbol{z}$, where Q is an orthogonal matrix given by $\begin{bmatrix} q^{\frac{1}{2}}\boldsymbol{1}_q \\ P \end{bmatrix}$, and $\boldsymbol{1}_q$ is a $q \times 1$ vector with all elements unity, leads to $u = 0$ and $\boldsymbol{v} = P\boldsymbol{z}$, with domain of v given by $\{\boldsymbol{v} = (v_1, v_2, \ldots, v_{(q-1)})^T : \boldsymbol{v}^T\boldsymbol{v} \leq 1\}$. One can easily express a Scheffé's model in terms of $\boldsymbol{v}$.

The problem of determining optimum designs in terms of $\boldsymbol{v}$ in its domain is a standard one in the context of response surface. Invariance structure, combined with the Loewner ordering (partial ordering of the information matrices), constitutes the Kiefer ordering, and this provides an effective tool in tackling optimal design problems of high dimension (*cf.* Pukelsheim, 1993). Using inverse transformation, it is easy to obtain the optimal design in terms of $\boldsymbol{z}$, and hence in terms of $\boldsymbol{x}$. Further, the optimum design in terms of $\boldsymbol{x}$ may not include the vertex points of the simplex, and, therefore, it will be different from the standard optimum design obtained over the whole simplex.

When Scheffé's first order mixture model is considered in the restricted space, the model in terms of $\boldsymbol{v}$ is also of first order, and to construct Kiefer optimal design on the experimental domain $\boldsymbol{v}^T\boldsymbol{v} \leq 1$ one needs to vary each of the $k = q-1$ components of $\boldsymbol{v}$ on the two levels $\pm\ k^{-1/2}$ only. The design that assigns uniform weight to each of the 2^k vertices of $[-k^{-1/2}, k^{-1/2}]^k$ is the complete factorial design 2^k, and its optimality is established through the following lemma (*cf.* Pukelsheim, 1993):

Lemma 1: A first order design $D(n \times k)$ with k components is optimum in the sense of Kiefer ordering if $D^TD \ \propto\ I_k$.

Examples of first order optimal designs for the restricted region are obtained by exploiting the Kiefer optimal first order designs on the $\boldsymbol{v}$ − space and choice of H as follows:

(i) For $q = 2$, $v \in [-1, 1]$, and the Kiefer optimal design assigns equal mass, namely $\frac{1}{2}$, to the two extreme points $v = -1$ and $v = 1$. Accordingly, the optimal design on the original restricted domain has the support points

(a) $\left(\frac{\sqrt{3}+\sqrt{2}}{2\sqrt{3}}, \frac{\sqrt{3}-\sqrt{2}}{2\sqrt{3}}\right)$ and $\left(\frac{\sqrt{3}-\sqrt{2}}{2\sqrt{3}}, \frac{\sqrt{3}+\sqrt{2}}{2\sqrt{3}}\right)$ when $H = 3^{-1/2}\ I_2$,

(b) $\left(\frac{\sqrt{2}+1}{2\sqrt{2}}, \frac{\sqrt{2}-1}{2\sqrt{2}}\right)$ and $\left(\frac{\sqrt{2}-1}{2\sqrt{2}}, \frac{\sqrt{2}+1}{2\sqrt{2}}\right)$ when $H = 2^{-1}\ I_2$,

In general, for $q = 2$, Lemma1 establishes the Keifer optimality of the designs obtained for all H of the form $H = hI_q$, when $h \leq 2^{-\frac{1}{2}}$.

It is to be noted that for $H = 2^{-1/2}\ I_2$ the points in the experimental region Ξ_0 are restricted by $x_1(x_1-1) \leq 0$, which leads to the optimum design in the restricted space to have support points at $(1, 0)$ and $(0, 1)$ with equal masses. This is also the case in the unrestricted case.. Draper and Pukelsheim (1999) has already established the Kiefer optimality of this design in their ingenious way.

(ii) For $q = 3$, the Kiefer optimal design in the $\boldsymbol{v}$ − space has the supports $\left(-\frac{\sqrt{3}}{2},\ \frac{1}{2}\right)$, $\left(\frac{\sqrt{3}}{2}, \frac{1}{2}\right)$, $(0,\ -1)$, which, on inverse transformation, gives the support points of the optimal design in the restricted space as $(1/6, 1/6, 2/3)$, $(2/3, 1/6, 1/6)$ and $(1/6, 2/3, 1/6)$ when $H = 6^{-1/2}\ I_3$, and $P = \begin{pmatrix} -\frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{6}} & -\frac{2}{\sqrt{6}} & \frac{1}{\sqrt{6}} \end{pmatrix}$. This design is an axial design as noted in Figure 3 below.

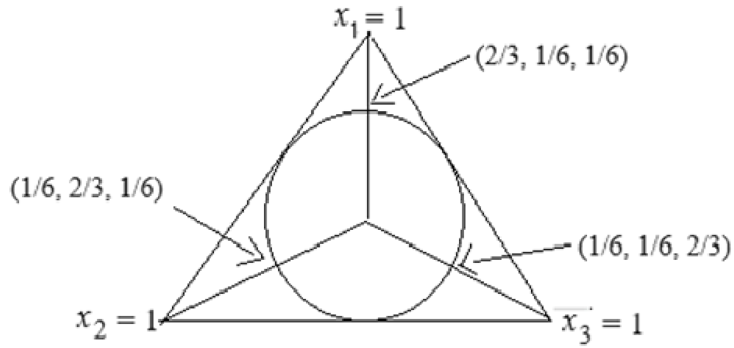


Figure 3: Support points of the optimum design in the case of 3-component mixture with ellipsoidal experimental region

Mandal *et al.* (2015) established that a design with any three points on the circumference of the circle in Figure 3, which form an equilateral triangle, is Kiefer-optimal. Thus, though the design indicated in Figure 3 is an axial design, a Kiefer optimal design is not necessarily so.

For Scheffé's quadratic mixture model, using the natural constraint $\sum_{i=1}^{k} x_i = 1$, it is possible to have a Kronecker product representation of the model, *viz.* $\eta_{\boldsymbol{x}} = (\boldsymbol{x} \otimes \boldsymbol{x})^T \boldsymbol{\beta}^*$, which makes it easier to represent the model as a quadratic model in $\boldsymbol{v}$ with its parameter vector, say τ^*, having a linear relationship with $\boldsymbol{\beta}^*$. As such, a design for estimating τ^* with Loewner Order dominance will also have Loewner Order dominance for τ^*. Pal *et al.* (2015) exploited this to obtain a Kiefer optimal design in the ellipsoidal region under Scheffé's quadratic mixture model.

Consider the central composite design (CCD) ξ^* in the $\boldsymbol{v}$ -space $\{\boldsymbol{v} : \boldsymbol{v}^T\boldsymbol{v} \leq 1\}$, which is a mixture of three blocks of designs, *viz.*

(i) cubes ξ_c , where ξ_c is a regular 2^{k-r} fraction of the full factorial design (with levels $\pm 1/\sqrt{k}$), of resolution V. (For $k \leq 5$, we have to take 2^k full factorial design);

(ii) stars ξ_s, where ξ_s is a set of star points of the form $(\pm 1, 0, 0, ..., 0), (0, \pm 1, 0, ..., 0), ..., (0, 0, ..., \pm 1)$;

(iii) centre points : $\xi_0 = \{\boldsymbol{v} : \boldsymbol{v}^T\boldsymbol{v} = 0\}$,

and ξ^* is defined as

$$\xi^* = (1 - \alpha)\xi_0 + \alpha\tilde{\xi}, \tag{2}$$

where $\tilde{\xi} = \frac{n_c\xi_c + n_s\xi_s}{n}$, $n_c = k^2, n_s = 2^{k-r}$, $n = 2^{k-r}n_c + 2kn_s, 0 < \alpha < 1$ Mandal *et al.* (2015) proved the following Theorem:

Theorem 1: The class of central composite designs (CCD), given by (2), is complete in the sense that given any design ξ, there is always a CCD of the form ξ^* given by (2) which is better in terms of

(i) Kiefer ordering

(ii) ϕ-optimality, provided it is invariant with respect to orthogonal transformation.

Through inverse transformation, it is then easy to conclude that the Kiefer optimal design for parameter estimation in Scheffé's quadratic mixture model in the ellipsoidal experimental region is obtained from a CCD, which is Kiefer optimal for the model in terms of $\boldsymbol{v}$.

For a 3-component mixture, ξ^* is obtained from the following blocks of designs:

(i) 4 star points: $(\pm 1, 0), (0, \pm 1)$

(ii) 2^2 factorial design points: $\frac{1}{\sqrt{2}}(-1,1), \frac{1}{\sqrt{2}}(-1,1), \frac{1}{\sqrt{2}}(1,-1), \frac{1}{\sqrt{2}}(1,1)$

(iii) centre point: $(0,0)$.

Then, for $H = \sqrt{6}I_3$, the optimal design in the restricted experimental region has the supports

(1) $\left(\frac{1}{3}+\frac{1}{2\sqrt{3}}, \frac{1}{3}, \frac{1}{3}-\frac{1}{2\sqrt{3}}\right)$; (2) $\left(\frac{1}{3}-\frac{1}{2\sqrt{3}}, \frac{1}{3}, \frac{1}{3}+\frac{1}{2\sqrt{3}}\right)$; (3) $\left(\frac{1}{6}, \frac{2}{3}, \frac{1}{6}\right)$;

(4) $\left(\frac{1}{2}, 0, \frac{1}{2}\right)$; (5) $\left(\frac{1}{3}+\frac{1}{2\sqrt{6}}-\frac{1}{6\sqrt{2}}, \frac{1}{3}+\frac{1}{3\sqrt{2}}, \frac{1}{3}-\frac{1}{2\sqrt{6}}-\frac{1}{6\sqrt{2}}\right)$;

(6) $\left(\frac{1}{3}-\frac{1}{2\sqrt{6}}-\frac{1}{6\sqrt{2}}, \frac{1}{3}+\frac{1}{3\sqrt{2}}, \frac{1}{3}+\frac{1}{2\sqrt{6}}-\frac{1}{6\sqrt{2}}\right)$;

(7) $\left(\frac{1}{3}+\frac{1}{2\sqrt{6}}+\frac{1}{6\sqrt{2}}, \frac{1}{3}-\frac{1}{3\sqrt{2}}, \frac{1}{3}-\frac{1}{2\sqrt{6}}+\frac{1}{6\sqrt{2}}\right)$;

(8) $\left(\frac{1}{3}-\frac{1}{2\sqrt{6}}-\frac{1}{6\sqrt{2}}, \frac{1}{3}+\frac{1}{3\sqrt{2}}, \frac{1}{3}+\frac{1}{2\sqrt{6}}-\frac{1}{6\sqrt{2}}\right)$;

(9) $\left(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}\right)$.

which are presented in Figure 4.

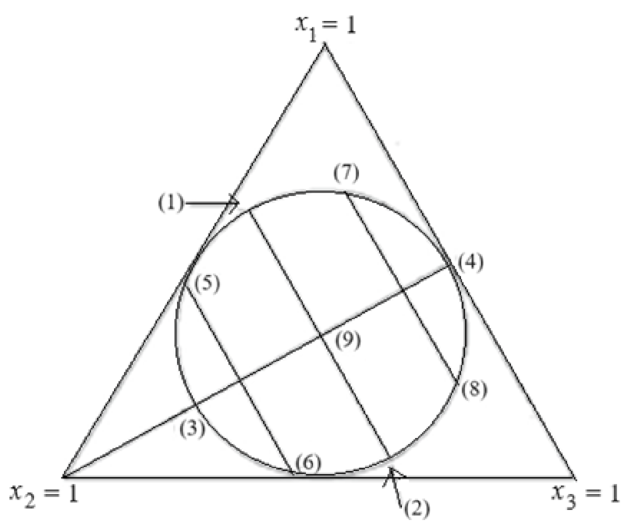


Figure 4: Support points of a Kiefer optimal design for the quadratic mixture model under ellipsoidal experimental region

It is noted that the Kiefer optimal design has 8 support points in the interior of the simplex, including the centroid, given by (9), and one on an edge, namely (4).

4. A restricted region in the form of a simplex within the unrestricted simplex

Mandal and Pal (2017) investigated the Kiefer optimal design when the experimental region is given by the form

$$\Xi_1 = \left\{ \boldsymbol{x} = (x_1, x_2, \ldots, x_q)^T : \frac{1}{q} - \frac{h}{q-1} \leq x_i \leq \frac{1}{q} + \frac{h}{q-1}, 1 \leq i \leq q, \sum_{i=1}^{q} x_i = 1 \right\},$$

where $h \in \left(0, \frac{q-1}{q}\right)$.

It is noteworthy that the centroid of the restricted region coincides with that of the simplex, *viz.* $\boldsymbol{x}_0 = \left(\frac{1}{q}, \frac{1}{q}, \ldots, \frac{1}{q}\right)^T$.

The transformation $\boldsymbol{x} \rightarrow \boldsymbol{z} = \frac{q-1}{qh}[\boldsymbol{x} - (\boldsymbol{x}_0 - \frac{h}{q-1}\mathbf{1}_q)]$ transforms the experimental region to

$$\Xi_{\boldsymbol{z}} = \left\{ \boldsymbol{z} = (z_1, z_2, \ldots z_q)^T : z_i \in [0.1], \ i = 1\,(1)\,q, \ \sum_{i=1}^{q} z_i = 1 \right\}, \tag{3}$$

which is same as the unrestricted experimental region Ξ.

The Kiefer optimal design in the permutation invariant class for estimation of the parameters of a first-degree or second-degree model, with unrestricted experimental region, is available in literature (Draper and Pukelsheim, 1999). This leads to the Kiefer optimal design for parameter estimation of the model in the restricted region owing to the 1:1 relation between $\boldsymbol{x}$ and $\boldsymbol{z}$.

For $q = 3$, the Kiefer optimal design in the $\boldsymbol{z}$-space has support points $(1, 0, 0)$, $(0, 1, 0)$ and $(0, 0, 1)$ for Scheffé's linear homogeneous model, and $(1, 0, 0)$, $(0, 1, 0)$, $(0, 0, 1)$, $(1/2, 1/2, 0)$, $(1/2, 0, 1/2)$ and $(0, 1/2, 1/2)$ for Scheffé's quadratic mixture model. This enables to find the support points of the Kiefer optimal design in the restricted $\boldsymbol{x}$-space as shown in Figure 5 for Scheffé's first order mixture model, and Figure 6 for Scheffé's quadratic mixture model. The points marked by alphabets in parentheses denote the support points.

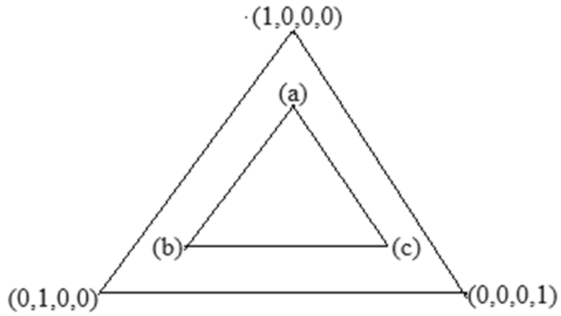
Remark: As the experimental region is well within the simplex, the vertex points of the simplex can never be included in a design.

5. Cuboidal experimental region in the simplex

A q-dimensional hypercube often defines the experimental region in industrial experimentation. In case of a cuboidal region, no result has been established so far that could help in finding the Kiefer optimal design. In view of that, Mandal and Pal (2017) attempted to find the D-optimal design for parameter estimation in the linear, homogeneous and quadratic mixture models due to Scheffé in the cuboidal region.

The restricted region within the simplex is defined by

$$\Xi_2 = \left\{ (x_1, \ldots, x_q)^T : 0 \leq x_{i0} - h_i \leq x_i \leq x_{i0} + h_i, \ \sum_{i=1}^{q} x_i = 1 \right\},$$

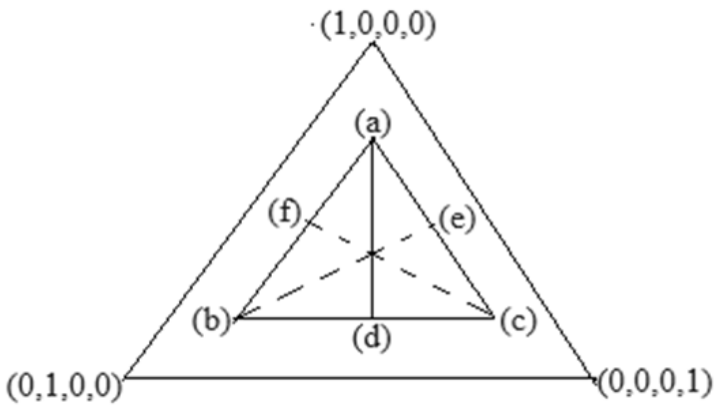


$$(a) = \left(\frac{1}{3} + \frac{4}{9h}, \frac{1}{3} - \frac{2}{9h}, \frac{1}{3} - \frac{2}{9h}\right)$$

$$(b) = \left(\frac{1}{3} - \frac{2}{9h}, \frac{1}{3} + \frac{4}{9h}, \frac{1}{3} - \frac{2}{9h}\right)$$

$$(c) = \left(\frac{1}{3} - \frac{2}{9h}, \frac{1}{3} - \frac{2}{9h}, \frac{1}{3} + \frac{4}{9h}\right)$$

Figure 5: Support points of a Kiefer optimal design for the first-order mixture model in the experimental region Ξ_1



$$(a) \left(\frac{1}{3} + h, \frac{1}{3} - \frac{h}{2}, \frac{1}{3} - \frac{h}{2}\right); (b) \left(\frac{1}{3} - \frac{h}{2}, \frac{1}{3} + h, \frac{1}{3} - \frac{h}{2}\right); (c) \left(\frac{1}{3} - \frac{h}{2}, \frac{1}{3} - \frac{h}{2}, \frac{1}{3} + h\right)$$

$$(d) \left(\frac{1}{3} + \frac{h}{4}, \frac{1}{3} + \frac{h}{4}, \frac{1}{3} - \frac{h}{2}\right); (e) \left(\frac{1}{3} + \frac{h}{4}, \frac{1}{3} - \frac{h}{2}, \frac{1}{3} + \frac{h}{4}\right); (f) \left(\frac{1}{3} - \frac{h}{2}, \frac{1}{3} + \frac{h}{4}, \frac{1}{3} + \frac{h}{4}\right)$$

Figure 6: Support points of a Kiefer optimal design for the quadratic mixture model in the experimental region Ξ_1

where $\boldsymbol{x}_0 = (x_{10}, \ldots, x_{q0})^T$ is the centre of Ξ_2, $h_i \leq \min\left[x_{i0}, 1 - x_{i0}\right] \;\; \forall i = 1\,(1)\,q$, and it is assumed that $\boldsymbol{x}_0$ is the centroid of the simplex .

A transformation $\boldsymbol{x} \rightarrow \boldsymbol{z} = H^{-1}(\boldsymbol{x} - \boldsymbol{x}_0)$, where $H = Diag(h_1,\ h_2, \ldots, h_q)$, along with the natural constraint $\sum_{i=1}^{q} x_i = 1$, gives

$-1 \leq z_i \leq 1$, for $i = 1\,(1)\,q$, $\quad \sum_{i=1}^{q} h_i z_i = 0.$

For $H \propto I_q$, or $h_i = h$ for all i, the $\boldsymbol{z}$ – space reduces to

$$\Xi_{\boldsymbol{z}} = \left\{ \boldsymbol{z} = (z_1, z_2, \ldots\ z_q)^T : -1 \leq z_i \leq 1, ,\ i = 1\,(1)\,q,\ \sum_{i=1}^{q} z_i = 0 \right\}.$$

A further orthogonal transformation $\boldsymbol{z} \rightarrow \begin{bmatrix} u \\ \boldsymbol{v}^{(q-1)\times 1} \end{bmatrix} = \begin{bmatrix} \sqrt{q}\mathbf{1}_q \\ P \end{bmatrix} \boldsymbol{z}$, gives $u = 0$, and the range of values of v_i as $-c \leq v_i \leq c$, where $\boldsymbol{v} = (v_1, v_2, \ldots\ v_{q-1})$ and

$$c \leq c^* = \min_{1 \leq i \leq q} x_{i0} \left[\frac{1}{h_i^2} - \frac{1}{a - h_i^2} \right]^{1/2}, a = \sum_{i=1}^{q} h_i^2$$

(vide Cornell (2002), pp. 122). c^* gives the greatest possible distance from $\boldsymbol{x} = \boldsymbol{x}_0$ to the closest boundary opposite the vertex $x_i = 1$.

Expressing the Scheffé's response model in terms of $\boldsymbol{v}$, the problem of determining the D-optimum design in the domain $-c \leq v_i \leq c,\ i = 1\,(1)\,(q-1)$, is a standard one in the context of response surface and the results are well known (*cf.* Pukelsheim, 1993). Mandal and Pal (2017) made use of this to find the D-optimum design for parameter estimation in the model in $\boldsymbol{x}$ with cuboidal experimental region.

For Scheffé's first-degree model in $\boldsymbol{x}$, the model in terms of $\boldsymbol{v}$ is also a first-degree model with its parameters sharing a 1:1 relationship with the parameters of the model in $\boldsymbol{x}$, and the restricted $\boldsymbol{x}$ -space Ξ_2 is permutation invariant. Hence, If $\xi_{\boldsymbol{x}}$ is a design in Ξ_2 corresponding to a design $\xi_{\boldsymbol{v}}$ in the $\boldsymbol{v}$-space, and, if $\xi_{\boldsymbol{v}}$ is D-optimal in the $\boldsymbol{v}$–space, then $\xi_{\boldsymbol{x}}$ will also be D-optimal in Ξ_2.

For $q = 2$ and $h_i = h$ for all i, v is a single variable in the interval $[-c, c]$. In this case, the D-optimal design in the v – space assigns mass $1/2$ at each of the values $-c$ and $+c$. Accordingly, the D-optimal design in the restricted space of $\boldsymbol{x}$ puts equal masses at $(x_{01} - \frac{hc}{\sqrt{2}},\ x_{02} + \frac{hc}{\sqrt{2}})$ and $(x_{01} + \frac{hc}{\sqrt{2}},\ x_{02} - \frac{hc}{\sqrt{2}})$.

For $q = 3$, the D-optimal design assigns equal masses at its support points $(\pm c, \pm c)$. Reverse transformation gives the support points of the D-optimal design with equal masses in the restricted $\boldsymbol{x}$ – space as

$$\left(x_{01} + \frac{\left(\sqrt{3} - 1\right) hc}{\sqrt{6}}, x_{02} + \frac{2hc}{\sqrt{6}}, x_{03} - \frac{\left(\sqrt{3} + 1\right) hc}{\sqrt{6}} \right)$$

$$\left(x_{01} + \frac{\left(\sqrt{3} + 1\right) hc}{\sqrt{6}}, x_{02} - \frac{2hc}{\sqrt{6}}, x_{03} - \frac{\left(\sqrt{3} - 1\right) hc}{\sqrt{6}} \right)$$

$$\left(x_{01} - \frac{\left(\sqrt{3}+1\right)hc}{\sqrt{6}}, x_{02} + \frac{2hc}{\sqrt{6}}, x_{03} + \frac{\left(\sqrt{3}-1\right)hc}{\sqrt{6}}\right)$$

$$\left(x_{01} - \frac{\left(\sqrt{3}-1\right)hc}{\sqrt{6}}, x_{02} - \frac{2hc}{\sqrt{6}}, x_{03} + \frac{\left(\sqrt{3}+1\right)hc}{\sqrt{6}}\right)$$

These points lie within the cuboidal region Ξ_2.

Remark: Different choices of H lead to different optimal designs in the restricted space.

In the quadratic response model due to Scheffé, using similar transformations [$\boldsymbol{x} \to \boldsymbol{z} \to (0, \boldsymbol{v})$], and observing that there is a 1:1 relation between the parameters of the model in terms of $\boldsymbol{x}$ and that in terms of $\boldsymbol{v}$, the D-optimal design in the $\boldsymbol{v}$–space leads to the D-optimal design in the restricted $\boldsymbol{x}$–space through reverse transformation.

From Mandal (1989), the support points of the D-optimal design in the $\boldsymbol{v}$–space is obtained from the following result:

Theorem 2: D-optimum design in the $\boldsymbol{v}$–space is supported on the lattice of points with coordinates only 0 or $\pm c$.

For the case of 3-component mixture, the support points of the D-optimum design in the $\boldsymbol{v}$–space are the points $(0,0), (\pm c, 0), (0, \pm c), (\pm c, \pm c)$.

Reverse transformation gives the support points of the D-optimum design in the restricted $\boldsymbol{x}$–space as

(i) (x_{01}, x_{02}, x_{03}) with mass 0;

(ii) $\left(x_{01} - \frac{hc}{\sqrt{2}}, x_{02}, x_{03} + \frac{hc}{\sqrt{2}}\right)$, $\left(x_{01} + \frac{hc}{\sqrt{2}}, x_{02}, x_{03} - \frac{hc}{\sqrt{2}}\right)$,

$$\left(x_{01} - \frac{hc}{\sqrt{6}}, x_{02} - \frac{2hc}{\sqrt{6}}, x_{03} + \frac{hc}{\sqrt{6}}\right), \left(x_{01} + \frac{hc}{\sqrt{6}}, x_{02} + \frac{2hc}{\sqrt{6}}, x_{03} - \frac{hc}{\sqrt{6}}\right)$$

each with mass 0.1325;

(iii) $\left(x_{01} + \frac{\left(\sqrt{3}-1\right)hc}{\sqrt{6}}, x_{02} + \frac{2hc}{\sqrt{6}}, x_{03} - \frac{\left(\sqrt{3}+1\right)hc}{\sqrt{6}}\right)$, $\left(x_{01} + \frac{\left(\sqrt{3}+1\right)hc}{\sqrt{6}}, x_{02} - \frac{2hc}{\sqrt{6}}, x_{03} - \frac{\left(\sqrt{3}-1\right)hc}{\sqrt{6}}\right)$, $\left(x_{01} - \frac{\left(\sqrt{3}+1\right)hc}{\sqrt{6}}, x_{02} + \frac{2hc}{\sqrt{6}}, x_{03} + \frac{\left(\sqrt{3}-1\right)hc}{\sqrt{6}}\right)$, $\left(x_{01} - \frac{\left(\sqrt{3}-1\right)hc}{\sqrt{6}}, x_{02} - \frac{2hc}{\sqrt{6}}, x_{03} + \frac{\left(\sqrt{3}+1\right)hc}{\sqrt{6}}\right)$

each with mass 0.1175.

6. Concluding remarks

The restricted experimental regions reviewed in this paper are proper subspaces of the simplex, and have suitable permutation invariance property. As such, it has been possible to characterize the optimal designs for Scheffé's linear and quadratic mixture models. Though the derivations are non-trivial, the tools used in earlier studies have been exploited to find the optimum designs.

In case of the absence of symmetry and invariance, it would be very difficult to obtain the optimum designs. This is the case if (i) the restricted region has its centre/centroid different from the centroid of the unrestricted simplex, or (ii) $H \propto I_q$ does not hold for the ellipsoidal/cuboid region. These remain as open problems which perhaps would be very challenging to tackle analytically.

Acknowledgement

The author thanks the anonymous reviewer for the fruitful comments, which helped to improve the presentation of the paper.

Conflict of interest

The authors do not have any financial or non-financial conflict of interest to declare for the research work included in this article.

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