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OPTIMAL DESIGNS FOR AN ADDITIVE QUADRATIC MIXTURE MODEL INVOLVING THE AMOUNT OF MIXTURE

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Abstract: This paper is concerned with D- and A-optimal designs for a quadratic additive model for experiments with mixtures, in which the response depends not only on the relative proportions but also on the actual amounts of the mixture components. It is found that the origin and vertices of the simplex are support points of these optimal designs, and when the number of mixture components increases, other support points shift gradually from barycentres of depth 1 to barycentres of higher depths. It is shown that the D-optimal designs have high efficiency in terms of A-optimality, and *vice versa*.

Key words and phrases: A-optimal design, additive model, D-optimal design, experiments with mixtures, mixture amount.

1. Introduction

Regression models for experiments with mixtures (Cornell (2002) and Chan (2000)) can be classified according to whether the response depends only on the relative proportions of the mixture components but not the actual amount of the mixture, or depends on both. The first type of model is called *A mixture model*, an example of which is the quality of a blend of wine which depends only on the composition of ingredients in the blend but not the actual quantity of wine in the bottle. An example of the second type looks at the effect of a fertilizer on a crop which depends not only on the composition but also the total amount of the fertilizer applied.

Let $a_i \geq 0$, $i = 1, \ldots, q$, be the actual amount of the i^{th} component in a mixture, and $a_1 + \cdots + a_q \leq A$, where A is a possible maximum total amount of the mixture. Let $x_i = a_i/A$, $i = 1, \ldots, q$, be the proportion of the i^{th} component relative to the maximum total amount A. Thus $\mathbf{x} = (x_1, \ldots, x_q)'$ belongs to the q-dimensional simplex $S^q = \{\mathbf{x} \in R^q : \ x_1 + \cdots + x_q \leq 1, \ \ x_i \geq 0, \ 1 \leq i \leq q\}$. Consider the model defined on S^q, in which the expected response at $\mathbf{x}$ is

$$\zeta_{DW2}(\mathbf{x}) = \beta_0 + \sum_{1\leq i\leq q} \beta_i x_i + \sum_{1\leq i\leq q} \theta_i x_i(1 - x_i). \tag{1.1}$$

In the spirit of Hilgers and Bauer (1995) and Heiligers and Hilgers (2003), we call the model (1.1), with the design space S^q, a component amount model; compare Piepel and Cornell (1985). Note that the form of $\zeta_{DW2}(\cdot)$ remains the same when A is replaced by another possible maximum total amount A^*. To see this, let $y_i = a_i/A^*$ and $\mathbf{y} = (y_1, \ldots, y_q)'$. From $\zeta_{DW2}(\mathbf{x}) = \zeta_{DW2}(\mathbf{y})$, it is easy to see that $\zeta_{DW2}(\mathbf{y})$ can be expressed in the same form as on the right hand side of (1.1) with x_i's replaced by y_i's. When $x_1 + \cdots + x_q = 1$, the design space will be the $(q-1)$-dimensional simplex $S^{q-1} = \{\mathbf{x} \in R^q: \ x_1 + \cdots + x_q = 1, \ \ x_i \geq 0, \ 1 \leq i \leq q\}$, and the constant β_0 on the right hand side of (1.1) can be absorbed into the other β_is (Cornell (2002), Section 2.2), forming the Darroch and Waller (1985) quadratic mixture model with $2q$ terms. The Darroch and Waller quadratic mixture model is additive in $x_1, \ldots, x_q$, has fewer terms than the Scheffé (1958) quadratic mixture model (which has $q(q+1)/2$ terms) when $q \geq 4$, but often fits data well (Chan (2000, Section 6)). Results on optimal designs for this model are available (Zhang and Guan (1992), Chan, Guan and Zhang (1998) and Chan, Meng and Jiang (1998)).

Few results are available on optimal designs for component amount models, other than Hilgers and Bauer (1995) and Heiligers and Hilgers (2003). The purpose of the present paper is to obtain optimal designs for the model in (1.1). Section 2 gives analytic results for D-optimal designs for $q = 4$ and $q \geq 8$, and A-optimal designs for $8 \leq q \leq 21$ and $q \geq 26$. For other values of q, approximately optimal designs are found by numerical searching using the computing package MATLAB. The origin and some vertices of S^q are support points in all cases. Some points on the edges of S^q are also support points for D-optimal design when $q = 3$, and A-optimal design when $3 \leq q \leq 7$. The results on D-optimality agree with the numerical findings for $q \leq 20$ in Heiligers and Hilgers (2003, p.723). Proofs of results are given in the Appendix.

2. Main Results

For $\delta \geq 0$, let $S_\delta^{q-1} = \{\mathbf{x} \in R^q: \ x_1 + \cdots + x_q = \delta, \ x_i \geq 0, \ 1 \leq i \leq q\}$. Denote S_1^{q-1} by S^{q-1}. A point $\mathbf{x} = (x_1, \ldots, x_q)' \in S_\delta^{q-1}$ is called a barycentre of depth $j-1$, $1 \leq j \leq q$, of S_δ^{q-1} if j of its q coordinates are equal to δ/j and the remaining coordinates are zero (Galil and Kiefer (1977)). Denote the collection of all barycentres of depth $j-1$ of S^{q-1} by J_j. In what follows, if δ is not mentioned, barycentres will refer to barycentres of S^{q-1}. For convenience, denote the binomial coefficient $q!/(j!\,(q-j)!)$ by $C(q,j)$. For any integers $0 < i_1 < i_2 < \cdots \leq q$, denote by $\boldsymbol{\xi}_{0,i_1,i_2,\ldots}$ a design in which a weight r_0 is assigned to the origin $\mathbf{0} = (0, \ldots, 0)' \in S^q$, a weight r_j is assigned to each point in J_j $(j = i_1, i_2, \ldots)$, where $C(q,0)r_0 + C(q,i_1)r_{i_1} + C(q,i_2)r_{i_2} + \cdots = 1$.

In the following, optimality will refer to optimality of design for the model $\zeta_{DW2}(\mathbf{x})$ in (1.1) defined on the design space S^q.

Theorem 2.1. *When $q = 4$, the design $\boldsymbol{\xi}_{0,1,i}$ with $i = 2$ and r_0, r_1, r_i defined by*

$$r_0 = 1/(2q+1), \quad C(q,1)r_1 = q/(2q+1), \quad C(q,i)r_i = q/(2q+1) \qquad (2.1)$$

is D-optimal.

Theorem 2.2. *When $q \geq 8$, the design $\boldsymbol{\xi}_{0,1,i}$ with $i = 3$ and r_0, r_1, r_i defined by* (2.1) *is D-optimal.*

When $q = 3$, it is verified numerically using MATLAB that D-optimality is achieved by the design which assigns a weight r_0 to the origin $(0,0,0)'$, a weight r_α to each of the points of the form $(\alpha,0,0)'$ (barycentres of depth 0 of S_α^{3-1}), and a weight r_i to each point in J_i $(i = 1,2)$, where $\alpha = 0.3825$, and the numerical values of r_0, r_α, r_1, r_2 are given in Table 1. This result agrees with that in Heiligers and Hilgers (2003, p.723).

For $5 \leq q \leq 7$, it is verified numerically that D-optimality is achieved by the design $\boldsymbol{\xi}_{0,1,2,3}$ with weights r_0, r_1, r_2, r_3 shown in Table 1.

In Table 1, for comparison, the values of $C(q,i)r_i$, $i = 0,1,2,3$, of the D-optimal designs (Zhang and Guan (1992)) for the Darroch and Waller quadratic mixture model defined on S^{q-1} are shown in smaller font in square brackets. The weights r_0 and r_α are not applicable to this model, since the origin and the points of the form $(\alpha,0,\ldots,0)'$, $0 < \alpha < 1$, do not belong to S^{q-1}. The same applies to Table 2. In Tables 1 and 2, "N.A." stands for "not applicable".

Table 1. D-optimal designs for the component amount model $\zeta_{DW2}(\mathbf{x})$ defined on S^q, and for the corresponding mixture model defined on S^{q-1}.

q	$C(q,0)r_0$	$C(q,1)r_1$	$C(q,2)r_2$	$C(q,3)r_3$	α	$C(q,1)r_\alpha$
3	0.1135 [N.A.]	0.4281 [1/2]	0.3777 [1/2]	0 [0]	0.3825 [N.A.]	0.0807 [N.A.]
4	$1/(2q+1)$ [N.A.]	$q/(2q+1)$ [1/2]	$q/(2q+1)$ [1/2]	0 [0]	N.A. [N.A.]	N.A. [N.A.]
5	0.0908 [N.A.]	0.4530 [0.4984]	0.4098 [0.4506]	0.0462 [0.0510]	N.A. [N.A.]	N.A [N.A.]
6	0.0769 [N.A.]	0.4577 [0.4959]	0.2528 [0.2753]	0.2125 [0.2288]	N.A. [N.A.]	N.A. [N.A.]
7	0.0666 [N.A.]	0.4644 [0.4977]	0.0850 [0.0877]	0.3842 [0.4146]	N.A. [N.A.]	N.A. [N.A.]
≥ 8	$1/(2q+1)$ [N.A.]	$q/(2q+1)$ [1/2]	0 [0]	$q/(2q+1)$ [1/2]	N.A. [N.A.]	N.A. [N.A.]

Table 2. A-optimal designs for the component amount model $\zeta_{DW2}(\mathbf{x})$ defined on S^q, and for the corresponding mixture model defined on S^{q-1}. Here $\gamma = 3 + 13\sqrt{q}$.

q	r_0	$C(q,1)r_1$	$C(q,2)r_2$	$C(q,3)r_3$	$C(q,4)r_4$	$\alpha(q)$	$C(q,1)r_{\alpha(q)}$
3	0.0119 [N.A.]	0.3378 [0.3923]	0.37075 [0.6077]	0 [0]	0 [0]	0.3508 [N.A.]	0.2798 [N.A.]
4	0.0187 [N.A.]	0.3630 [0.4142]	0.4339 [0.5858]	0 [0]	0 [0]	0.3279 [N.A.]	0.1845 [N.A.]
5	0.0003 [N.A.]	0.3211 [0.3496]	0 [0]	0.4716 [0.6504]	0 [0]	0.3213 [N.A.]	0.2070 [N.A.]
6	0.0515 [N.A.]	0.3346 [0.3496]	0 [0]	0.5517 [0.6504]	0 [0]	0.2954 [N.A.]	0.0622 [N.A.]
7	0.0473 [N.A.]	0.3446 [0.3496]	0 [0]	0.5507 [0.6504]	0 [0]	0.2760 [N.A.]	0.0582 [N.A.]
8	0.07350 [N.A.]	0.3534 [0.3814]	0 [0]	0.5731 [0.6186]	0 [0]	N.A. [N.A.]	N.A. [N.A.]
$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$
21	0.0473 [N.A.]	0.3820 [0.4010]	0 [0]	0.5706 [0.5990]	0 [0]	N.A. [N.A.]	N.A. [N.A.]
22	0.0462 [N.A.]	0.3603 [0.3946]	0 [0]	0.4478 [0.4687]	0.1457 [0.1367]	N.A. [N.A.]	N.A. [N.A.]
23	0.0453 [N.A.]	0.3657 [0.3881]	0 [0]	0.3209 [0.3328]	0.2681 [0.2791]	N.A. [N.A.]	N.A. [N.A.]
24	0.0444 [N.A.]	0.3770 [0.3818]	0 [0]	0.1778 [0.1974]	0.4008 [0.4208]	N.A. [N.A.]	N.A. [N.A.]
25	0.0435 [N.A.]	0.3721 [0.3769]	0 [0]	0.0611 [0.0676]	0.5233 [0.5565]	N.A. [N.A.]	N.A. [N.A.]
26	0.0428 [N.A.]	0.3572 [0.3732]	0 [0]	0 [0]	0.6000 [0.6268]	N.A. [N.A.]	N.A. [N.A.]
$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$
$\to\infty$	$3/\gamma$ [N.A.]	$5\sqrt{q}/\gamma$ [5/13]	0 [0]	0 [0]	$8\sqrt{q}/\gamma$ [8/13]	N.A. [N.A.]	N.A. [N.A.]

To express A-optimality for the model $\zeta_{DW2}(\mathbf{x})$, define r_0, r_1, r_i, $i \geq 2$, by

$$r_0 : C(q,1)r_1 : C(q,i)r_i = 1 : \alpha(q,i) : \beta(q,i), \tag{2.2}$$

$$\alpha(q,i) = \Big(q^2(2i^2 - 2i + 1)/((q+1)(i-1)^2)\Big)^{1/2}, \tag{2.3}$$

$$\beta(q,i) = \Big(i^3 q(qi-2i+1)C(q,i)/((q+1)(q-1)(i-1)^2 C(q-2,i-1))\Big)^{1/2}. \tag{2.4}$$

Theorem 2.3. *When $8 \leq q \leq 21$, the design $\boldsymbol{\xi}_{0,1,i}$ with $i = 3$ and r_0, r_1, r_i*

defined by (2.2)−(2.4) *is A-optimal.*

Theorem 2.4. *When $q \geq 26$, the design $\boldsymbol{\xi}_{0,1,i}$ with $i = 4$ and r_0, r_1, r_i defined by* (2.2)−(2.4) *is A-optimal.*

As for $q = 3, \ldots, 7$, numerical searching using MATLAB shows that A-optimality is achieved by the designs that assign a weight r_0 to the origin $(0, \ldots, 0)'$, a weight $r_{\alpha(q)}$ to each of the points of the form $(\alpha(q), 0, \ldots, 0)$ for a specific $\alpha(q) \in (0, 1)$, and a weight r_i to each point in J_i, where $i = 2$ when $q = 3, 4$, and $i = 3$ when $q = 5, 6, 7$. The numerical values of the r_i's, $\alpha(q)$'s and $r_{\alpha(q)}$'s, $q = 3, \ldots, 7$, are given in Table 2. In Table 2, for comparison, the values of $C(q, i)r_i$, $i = 1, 2, 3, 4$, of the A-optimal designs (Chan, Guan and Zhang (1998)) for the quadratic Darroch and Waller mixture model defined on S^{q-1} mixture are shown in smaller font in square brackets.

For the case $22 \leq q \leq 25$, it is shown numerically that the designs $\boldsymbol{\xi}_{0,1,3,4}$ with values of r_0, r_1, r_3, r_4 shown in Table 2 are A-optimal.

3. Discussion

The results in Section 2 show that for some values of q, points of the form $(\alpha, 0, \ldots, 0)$ $(0 < \alpha < 1)$ are support points for optimal designs. This does not contradict a result of Atwood (1969, pp.1573-1574) which states that only barycentres support optimal designs for n-tic polynomial mixture models defined on S^{q-1} on which the condition $x_1 + \cdots + x_q = 1$ is satisfied. However, in the component amount model $\zeta_{DW2}(\mathbf{x})$ in (1.1) defined on S^q, $x_1 + \cdots + x_q$ can take any value lying within 0 and 1. Atwood's argument shows that for any fixed $\delta > 0$ and for an n-tic polynomial model defined on S_δ^{q-1}, only barycentres of S_δ^{q-1} are possible support points for D- or A-optimal designs. Since $S^q = \cup_{\delta \in [0,1]} S_\delta^{q-1}$, it is possible that barycentres of some S_δ^{q-1}, $\delta \in (0, 1)$, are support points for a D- or A-optimal design for the model $\zeta_{DW2}(\mathbf{x})$ defined on S^q.

To compare the efficiency of designs, define the D-efficiency e_D of the design $\boldsymbol{\xi}$ relative to the design $\boldsymbol{\xi}_0$, and the A-efficiency e_A of $\boldsymbol{\xi}$ relative to $\boldsymbol{\xi}_0$ for the same regression model by

$$e_D = (\det M(\boldsymbol{\xi}) / \det M(\boldsymbol{\xi}_0))^{1/s}, \tag{3.1}$$

$$e_A = \operatorname{tr} M^{-1}(\boldsymbol{\xi}_0) / \operatorname{tr} M^{-1}(\boldsymbol{\xi}), \tag{3.2}$$

respectively, where s is the number of coefficients in the regression model. Hence the larger the value of e_D or e_A in (3.1) or (3.2), the more efficient the design $\boldsymbol{\xi}$ relative to $\boldsymbol{\xi}_0$, and if $\boldsymbol{\xi}_0$ is optimal, the largest possible value of e_D or e_A is 1. The e_D values of the A-optimal designs and the e_A values of the D-optimal designs for the model $\zeta_{DW2}(\mathbf{x})$ in (1.1) are computed. Table 3 which shows these values for $3 \le q \le 10$ and for $q \to \infty$ indicates that the D-optimal designs are very efficient in terms of A-optimality, and the A-optimal designs are very efficient in terms of D-optimality.

Table 3. D- and A-efficiencies of optimal designs for $\zeta_{DW2}(\mathbf{x})$.

q	e_D	e_A
3	0.962858	0.923901
4	0.967339	0.953793
5	0.923067	0.927907
6	0.953325	0.917221
7	0.963075	0.935227
8	0.971470	0.947673
9	0.972430	0.948973
10	0.972947	0.949354
$\to \infty$	$\to 1$	$\to 1$

Appendix.

In what follows, let $\mathbf{I}_a$ denote the $a \times a$ identity matrix, $\mathbf{1}_{a\times b}$ denote the $a \times b$ matrix of 1's, and $\mathbf{0}_{a\times b}$ denote the $a \times b$ matrix of 0's. Let M_i be a $C(q,i) \times q$ matrix such that the first i elements in the first row of M_i are 1, the remaining elements in the first row are 0, and the remaining $C(q,i) - 1$ rows of M_i are the different permutations of the first row according to lexicographical order. For the model $\zeta_{DW2}(\mathbf{x})$ in (1.1), it is straightforward to show that the model matrix generated by all points in J_1 is $(\mathbf{I}_q, \mathbf{0}_{q\times q})$ and, for any fixed integer $i = 2, \ldots, q$, the model matrix generated by all points in J_i is $(i^{-1}M_i, (i-1)i^{-2}M_i)$.

For the design $\boldsymbol{\xi}_{0,1,i}$, we require

$$r_0 + C(q,1)r_1 + C(q,i)r_i = 1, \tag{A.1}$$

and the moment matrix associated with $\boldsymbol{\xi}_{0,1,i}$ is given by

$$M(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,i}) = \begin{pmatrix} 1 & k_1\mathbf{1}_{1\times q} & k_2\mathbf{1}_{1\times q} \\ k_1\mathbf{1}_{q\times 1} & r_1\mathbf{I}_q + r_i\, i^{-2}\, M_i'M_i & (i-1)\, r_i\, i^{-3}\, M_i'M_i \\ k_2\mathbf{1}_{q\times 1} & (i-1)\, r_i\, i^{-3}\, M_i'M_i & (i-1)^2\, r_i\, i^{-4}\, M_i'M_i \end{pmatrix}, \tag{A.2}$$

where $k_1 = r_1 + r_i\, i^{-1}\, C(q-1, i-1)$, $k_2 = (i-1)\, r_i\, i^{-2}\, C(q-1, i-1)$, and $M_i'M_i = C(q-2, i-1)\mathbf{I}_q + C(q-2, i-2)\mathbf{1}_{q\times q}$. Applying a formula for the determinant of a partitioned matrix (Morrison (1976), Section 2.11) twice, we find that

$$\det M(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,i}) = (i-1)^{2q}\, i^{-4q} \det(M_i' M_i)\, r_0\, r_1^q\, r_i^q.$$

By the method of Lagrange multipliers, it can be shown that for a fixed i, and under the constraint (A.1), the only critical point of $\det M(\boldsymbol{\xi}_{0,1,i})$ is a maximum point attained at the r_0, r_1, r_i that satisfy (2.1).

As for A-optimality, it follows readily from Morrison (1976, Section 2.11) that the inverse of the moment matrix $M(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,i})$ in (A.2) is given by

$$M^{-1}(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,i})$$
$$= \begin{pmatrix} r_0^{-1} & -r_0^{-1}\,\mathbf{1}_{1\times q} & \mathbf{0}_{1\times q} \\ -r_0^{-1}\,\mathbf{1}_{q\times 1} & r_0^{-1}\,\mathbf{1}_{q\times q} + r_1^{-1}\,\mathbf{I}_q & -i(i-1)^{-1}\, r_1^{-1}\mathbf{I}_q \\ \mathbf{0}_{q\times 1} & -\,i(i-1)^{-1}\, r_1^{-1}\mathbf{I}_q & i^2(i-1)^{-2}(r_1^{-1}\,\mathbf{I}_q + i^2\, r_i^{-1}\,(M_i' M_i)^{-1}) \end{pmatrix},$$

where $(M_i' M_i)^{-1} = (\mathbf{I}_q - (i-1)i^{-1}(q-1)^{-1}\mathbf{1}_{q\times q})/C(q-2, i-1)$.

Consequently, we have

$$\operatorname{tr} M^{-1}(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,i}) = \frac{q+1}{r_0} + \frac{q(2i^2 - 2i + 1)}{(i-1)^2 r_1} + \frac{i^3 q(qi-2i+1)}{(i-1)^2(q-1)C(q-2, i-1) r_i}. \tag{A.3}$$

By the method of Lagrange multipliers, it can be shown that the only minimum point of $\operatorname{tr} M^{-1}(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,i})$ is attained at the r_0, r_1, r_i that satisfy (2.2)–(2.4).

If $\mathbf{x} = (x_1, \ldots, x_q)'$, let $\mathbf{f}(\mathbf{x}) = (1, x_1, \ldots, x_q, x_1(1-x_1), \ldots, x_q(1-x_q))'$. Since $S^q = \cup_{\delta\in[0,1]} S_\delta^{q-1}$, according to the well-known equivalence theorems for optimality (Kiefer (1974, 1975)), a design $\boldsymbol{\xi}$ is D-optimal and A-optimal for the model $\zeta_{DW2}(\mathbf{x})$ in (1.1) defined on S^q if and only if

$$f_D(\zeta_{DW2}, \boldsymbol{\xi}; \mathbf{x}) = (2q+1)^{-1}\mathbf{f}'(\mathbf{x})M^{-1}(\zeta_{DW2}, \boldsymbol{\xi})\mathbf{f}(\mathbf{x}) - 1 \le 0 \tag{A.4}$$

and

$$f_A(\zeta_{DW2}, \boldsymbol{\xi}; \mathbf{x}) = \mathbf{f}'(\mathbf{x})M^{-2}(\zeta_{DW2}, \boldsymbol{\xi})\mathbf{f}(\mathbf{x}) - \operatorname{tr} M^{-1}(\zeta_{DW2}, \boldsymbol{\xi}) \le 0, \tag{A.5}$$

respectively, for all $\mathbf{x} \in S_\delta^{q-1}$ and for all $0 \le \delta \le 1$. Furthermore, the second equality in (A.4) or in (A.5) occurs at all points in the support of a D- or A-optimal design.

Both $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ are fourth degree polynomials in $x_1, \ldots, x_q$, symmetric in any pair of coordinates, and approach infinity as any one of $x_1, \ldots, x_q$ approaches infinity. If $\mathbf{x} \in S^q$, $\mathbf{x}$ must be either the origin or belongs to S_δ^{q-1} for some $\delta \in (0, 1]$. Let $\mathbf{x} \in S_\delta^{q-1}$, where $\delta > 0$. Fix all but two of x_i, $i = 1, \ldots, q$, say x_1 and x_2. Then $x_2 = K - x_1$ for some constant K, and both $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ can be expressed as fourth degree polynomials of the single variable x_1. Thus each of $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ have at most three maximum points, two at the end points $(x_1 = 0, K)$ and one in the interior of the range $[0, K]$ of x_1. Since both $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ are symmetric in x_1 and x_2, interchanging the roles of x_1 and x_2 shows that both $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ attain their maximums either when $(x_1, x_2) = (0, K)$, $(x_1, x_2) = (K, 0)$, or $x_1 = x_2 = K/2$. Repeating the above with the roles of x_1, x_2 replaced by the other x_is, $i \neq 1, 2$, shows that both $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ attain their maxima when some or none of x_i is 0 and the non-zero x_i's, $i = 1, \ldots, q$, take equal values. In other words, if $\delta > 0$, $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ attain their maxima on S_δ^{q-1} only at barycentres of S_δ^{q-1}. If $\delta = 0$, S_δ^{q-1} reduces to the origin at which $f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ and $f_A(\zeta_{_{DW2}}, \boldsymbol{\xi}; \mathbf{x})$ may also attain a maximum. Thus, only barycentres of S_δ^{q-1} ($\delta \in [0, 1]$) are possible support points for D- or A-optimal designs for the model $\zeta_{_{DW2}}(\mathbf{x})$ defined on S^q.

Consequently, in order to prove that (A.4) or (A.5) is satisfied for all $\mathbf{x} \in S^q$, it suffices to prove that they are satisfied at $\mathbf{x} = \mathbf{0} = (0, \ldots, 0)'$ and at all barycentres of S_δ^{q-1} for all $\delta \in (0, 1]$.

To prove the results for D-optimality, we observe that for the design $\boldsymbol{\xi}_{0,1,i}$ in which the measures r_0, r_1, r_i are defined by (2.1), we have

$$\begin{aligned} f_D(\zeta_{_{DW2}}, \boldsymbol{\xi}_{0,1,2}; \mathbf{x}) = (2q+1)^{-1} \Bigg[& \frac{1}{r_0} \Big\{ 1 - 2\sum_{k=1}^{q} x_k + \Big(\sum_{k=1}^{q} x_k \Big)^2 \Big\} \\ & + \frac{1}{r_1} \Big\{ \sum_{k=1}^{q} (x_k^2 - \frac{2i}{i-1} x_k^2 (1 - x_k) + \frac{i^2}{(i-1)^2} x_k^2 (1 - x_k)^2) \Big\} \\ & + \frac{i^4}{r_i} \Big\{ \frac{(i-2)!\,(q-i-1)!}{(i-1)\,(q-2)!} \sum_{k=1}^{q} x_k^2 (1 - x_i)^2 \\ & \quad - \frac{(i-2)!\,(q-i-1)!}{i\,(q-1)!} \Big(\sum_{k=1}^{q} x_k (1 - x_k) \Big)^2 \Big\} \Bigg] - 1. \qquad \text{(A.6)} \end{aligned}$$

Proof of Theorem 2.1. Suppose that $q = 4$, $i = 2$, and $\boldsymbol{\xi}_{0,1,2}$ is the design in which the measures r_0, r_1, r_2 are defined by (2.1). We prove that (A.4) is satisfied.

When $\delta \in [0,1]$ and $\mathbf{x}$ is a barycentre of depth j of S_δ^{q-1}, it follows from (A.6) that $j^3 f_D(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,2}; \mathbf{x}) = \delta\left[2(8-j)\delta^3 + 4(-7+j)j\delta^2 + (13-j)j^2\delta - 2j^3\right] = \delta P_1(j,\delta)$, say. Hence $f_D(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,2}; \mathbf{x}) = 0$ when $\delta = 0$, that is, $\mathbf{x} = \mathbf{0}$. It is straightforward to show that for each $j = 1,2,3,4$, the function $P_1(j,\delta)$ attains its maximum in $\{\delta : \delta \in [0,1]\}$ at $\delta = 1$, and $P_1(j,1) = -3j^3 + 17j^2 - 30j + 16$. The last cubic polynomial in j equals 0 when $j = 1,2$, and is negative for all $j \geq 3$. Thus $f_D(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,2}; \mathbf{x}) \leq 0$ for all $\delta \in [0,1]$ and $j = 1,2,3,4$, and $f_D(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,2}; \mathbf{x}) = 0$ if and only if either $\mathbf{x} = \mathbf{0}$, or $\delta = 1$ and $j = 1,2$. Hence (A.4) is satisfied and the design $\boldsymbol{\xi}_{0,1,2}$ is D-optimal, and only the origin and points in J_1 and J_2 are possible support points. This proves Theorem 2.1.

Proof of Theorem 2.2. Suppose that $q \geq 8$, $i = 3$, and $\boldsymbol{\xi}_{0,1,3}$ is the design in which the measures r_0, r_1, r_3 are defined by (2.1). We show that (A.3) is satisfied.

When $\delta \in [0,1]$ and $\mathbf{x}$ is a barycentre of depth j of S_δ^{q-1}, it follows from (A.6) that $2j^3(q-3) f_D(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,3}; \mathbf{x}) = \delta[(-27+18q-9j)\delta^3 + (36-30q+18j)j\delta^2 - (15-14q+(15-2q)j)j^2\delta + 4(3-q)j^3] = \delta P_2(q,j,\delta)$, say. Hence $f_D(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,3}; \mathbf{x}) = 0$ when $\delta = 0$, or when $\mathbf{x} = \mathbf{0}$.

When $j = 1$, $P_2(q,j,\delta) = 2(\delta-1)[\delta(9-6q) + 2(q-3) + 9\delta^2(q-2)] \leq 0$, and the last equality holds if and only if $\delta = 1$.

When $j = 2$, $P_2(q,j,\delta) = \delta(96 - 180\delta + 144\delta^2 - 45\delta^3) + (-32 + 72\delta - 60\delta^2 + 16\delta^3)q \leq \delta(96 - 180\delta + 144\delta^2 - 45\delta^3) + (-32 + 72\delta - 60\delta^2 + 16\delta^3)8 < 0$ for all $\delta \in [0,1]$.

When $j = 3$, $P_2(q,j,\delta) = 18(\delta-1)\delta(6 - 4\delta + \delta^2)(q-3) \leq 0$, and the last equality holds when $\delta = 1$.

Now consider the case $4 \leq j \leq q$. The function $P_2(q,j,\delta)$ is a cubic polynomial in δ, and the coefficient of δ^3 is $(-27+18q-9j) \geq -27+18q-9q > 0$. We show that $\partial P_2(q,j,\delta)/\partial\delta = 3(-27+18q-9j)\delta^2 + 2(36j - 30jq + 18j^2)\delta + (-15j^2 + 14j^2q - 15j^3 + 2j^3q) = A\delta^2 + B\delta + C$, say, does not have a real zero, so that $P_2(q,j,\delta)$ is strictly increasing for all δ. From the equation $B^2 - 4AC = (16-12j)q^2 + (-24+30j+6j^2)q + (9-36j-9j^2) = 0$, q can be found in terms of j, and since $(16-12j) < -32 < 0$, $B^2 - 4AC \geq 0$ only for the range of values of q that lie between the two roots of the equation $B^2 - 4AC = 0$. The larger of these two roots is $q^* = \left(3j^2 + 15j - 12 + \sqrt{j(9j^3 - 6j^2 - 151j + 324)}\right)/(12j-16) < \left(3j^2 + 15j - 12 + (3j^2 - j - 15)\right)/(12j-16) < j$, where the last two inequalities hold because $j \geq 4$. Hence $B^2 - 4AC \geq 0$ only if $q < q^* < j$. Since

$q < q^* < j$ contradicts $4 \le j \le q$, we always have $B^2 - 4AC < 0$. Thus $\partial P_2(q,j,\delta)/\partial\delta \neq 0$ for all δ. It follows that $P_2(q,j,\delta)$ is strictly increasing in δ for all δ, and $P_2(q,j,\delta) \le P_2(q,j,1) = (j-1)(j-3)[(6q-9)-(2q+3)j] < 0$.

Therefore, $f_D(\zeta_{DW2}, \boldsymbol{\xi}_{0,1,3}; \mathbf{x}) \le 0$ at all barycentres of S_δ^{q-1} and for all $\delta \in [0,1]$, and the last equality holds if and only if either $\mathbf{x} = \mathbf{0}$ or $\mathbf{x} \in J_1 \cup J_3$. Thus (A.4) is satisfied, and Theorem 2.2 is proved.

To prove the results for A-optimality, we observe that if $\xi_{0,1,i}$ is the design in which the measures r_0, r_1, r_i are defined by (2.2)$-$(2.4), we have the following:

$$\begin{aligned}
&\mathbf{f}'(\mathbf{x})M^{-2}(\zeta_{DW2}, \boldsymbol{\xi})\mathbf{f}(\mathbf{x}) \\
&= \frac{q+1}{r_0^2} - 2\left(\frac{q+1}{r_0^2} + \frac{1}{r_0 r_1}\right)\sum_{k=1}^q x_k + \frac{2i}{(i-1)r_0 r_1}\sum_{k=1}^q x_k(1-x_k) \\
&\quad + \left(\frac{q+1}{r_0^2} + \frac{2}{r_0 r_1}\right)\left(\sum_{k=1}^q x_k\right)^2 - \frac{2i}{(i-1)r_0 r_1}\sum_{k=1}^q x_k \sum_{k=1}^q x_k(1-x_k) \\
&\quad + \sum_{k=1}^q \{a x_k^2 + 2b x_k^2(1-x_k) + 2c x_k(1-x_k) + d x_k^2(1-x_k)^2\} \\
&\quad + e\left\{\sum_{k=1}^q x_k(1-x_k)\right\}^2, \qquad \text{(A.7)}
\end{aligned}$$

where

$$\begin{aligned}
a &= \frac{1}{r_1^2} + \frac{i^2}{(i-1)^2 r_1^2}, \\
b &= \frac{-i}{(i-1)r_1^2} + \frac{-i^3}{(i-1)^3 r_1^2} + \frac{-i^5}{(i-1)^3 r_1 r_i C(q-2,i-1)}, \\
c &= \frac{i^4}{r_1 r_i (i-1)^2 (q-1) C(q-2,i-1)}. \\
d &= \frac{i^2}{(i-1)^2 r_1^2} + \left(\frac{i^2}{(i-1)^2 r_1} + \frac{i^4}{(i-1)^2 r_i C(q-2,i-1)}\right)^2, \\
e &= \frac{-2i^5}{(i-1)^3 r_1 r_i (q-1) C(q-2,i-1)} + \frac{-2i^7(q-1) + i^6 q(i-1)}{(i-1)^3 r_i^2 (q-1)^2 (C(q-2,i-1))^2}.
\end{aligned}$$

Proof of Theorem 2.3. Suppose $q \ge 8$, and $\xi_{0,1,3}$ is the design in which the measures r_0, r_1, r_3 are defined by (2.2)$-$(2.4).

Barycentres of S_δ^{q-1} are the only possible support points for an A-optimal design. Any barycentre of S_δ^{q-1} can be written as $\delta\mathbf{x}$, where $\mathbf{x}$ is a barycentre of S^{q-1}. Therefore, to prove Theorem 2.3 it suffices to show that (A.4) holds for all $\delta\mathbf{x}$, where $\delta \in [0,1]$ and $\mathbf{x}$ is a barycentre of S^{q-1}, and that equality in (A.4) holds if and only if either $\mathbf{x} = \mathbf{0}$, or $\delta = 1$ and $\mathbf{x} \in J_1 \cup J_2$.

Using (A.3) and (A.7), it follows, from straightforward but lengthy calculation using the computing package *Mathematica*, that if $\mathbf{x} \in J_j$, $1 \le j \le q$, $j^3 f_A(\zeta_{DW2}, \xi; \delta\mathbf{x}) = \delta[(K_1 + K_2 j)\delta^3 + (K_3 + K_4 j) j\delta^2 + \delta(K_5 + K_6 j) j^2 \delta + K_7 j^3] = \delta P_3(q, j, \delta)$, say, where $K_1, \ldots, K_7$ depend only on q. Hence $f_A(\zeta_{DW2}, \xi; \mathbf{0}) = 0$. Lengthy calculations show that $P_3(q,j,\delta) < P_3(q,j,1)$ for all $\delta \in [0,1)$. In $P_3(q,j,1)$, j can take any value (although $j-1$ has the geometric meaning of being the depth of barycentres only if j is an integer and $1 \le j \le q$). For any $q = 8, \ldots, 21$, it can be shown from lengthy calculations that $P_3(q,j,1) > 0$ when $j = 0$, $P_3(q,j,1) = 0$ when $j = 1, 3$, $P_3(q,j,1) < 0$ when $j = 2, 4$. Since $P_3(q,j,1)$ is a cubic polynomial in j, it has at most three zeros, and consequently $P_3(q,j,1)$ is positive for all $j \le 0$ and negative for all $j \ge 4$. Hence (A.5) is satisfied for all $\mathbf{x} \in S^q$, and the second equality in (A.5) holds if and only if $\delta = 1$ and $\mathbf{x} \in J_1 \cup J_3$. This proves that the design $\xi_{0,1,3}$ is A-optimal when $q = 8, \ldots, 21$, and only the origin and points in J_1 and J_3 are possible support points. This proves Theorem 2.3.

The proof of Theorem 2.4 is similar to that of Theorem 2.3 and is omitted.

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