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A NOTE ON OPTIMAL MIXTURE AND MIXTURE AMOUNT DESIGNS

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Abstract: In many applications of mixture experiments in medicine or biology, for example, not only the proportions of the involved mixture ingredients, but also their total amount is of particular interest. This calls for designs in mixture amount models, which are obtained from classical mixture setups by including terms capturing the total amount, simultaneously dropping the sidecondition on the proportions to sum up to one. While design optimality usually depends sensitively on the underlying model, we establish here a close relation between admissible mixture, and admissible mixture amount designs in additive and homogeneous models. This particularly allows to obtain D-, A- and V-optimal mixture amount from optimal mixture designs, and vice versa. We present some examples for Becker's and Scheffé's mixture models.

Key words and phrases: Admissibility, A-optimality, approximate design, Becker models, complete class, D-optimality, I-optimality, mixture amount experiments, mixture experiments, V-optimality, tic-type polynomials.

1. Introduction and Preliminaries

Many practical problems are associated with the investigation of mixture ingredients $t_1, \ldots, t_q$ of q factors, with $t_i \geq 0$, being further restricted by $\sum_{i=1}^{q} t_i = 1$. The definitive text Cornell (1990) lists numerous examples and provides a thorough discussion of both, theory and practice. Early seminal work was done by Scheffé (1958, 1963) who suggested (1958, p.347) and analyzed canonical model forms when the expected response $y = y(t)$ is a multiple polynomial of degree one, two, or three. Quenouille (1959) and others pointed out and illustrated that alternatives for describing mixture experiments are desirable, as polynomial models do not only have shortcomings for prediction purposes, but also do not satisfactorily account for mixture components which are inert or have additive effects on the response. Besides that, in polynomial mixture models the regression coefficients cannot be interpreted. For overcoming these disadvantages, Becker (1968) proposed additive mixture models constructed from functions which are homogeneous of the same degree (degree one, in his paper). The essential point in these classical mixture setups is that the response is supposed to depend only on the *proportions* of the involved ingredients, but not on their *total amount.* In many applications, however, the total amount is of particular interest, (due to toxic side effects in biology or medicine, for example). For investigating whether the blending properties of the ingredients change when the total amount of the mixture changes, so-called component amount models were suggested, which include terms capturing the total amount, see e.g., Piepel and Cornell (1985, 1994). Typically, the total amount of the individual components is zero in the placebo point or control test, cf. e.g., Piepel (1988). When normalizing the maximum total amount to one, the mixture ingredients $t_1, \dots, t_q \geq 0$ satisfy the restriction $\sum_{i=1}^{q} t_i \leq 1$ instead of summing to one.

Optimality properties of experimental designs usually depend sensitively on the underlying model. Actually, when taking the total amount of the mixture components into account, even the structure of optimal designs may change completely. The purpose of the present paper is to investigate in more detail the relation between optimal designs for mixture, and for component amount models. More precisely, we consider multifactor experiments, for q deterministic ingredients that are assumed to influence the response through the percentages (or proportions) in which they are blended together. For $i = 1, \dots, q$ let $t_i \in [0, 1]$ be the proportion of ingredient i in the mixture. As usual, we assemble the individual components to form the column vector $t = (t_1, \dots, t_q)'$ of experimental conditions, (the prime denotes transposition). In classical mixture experiments, the experimental domain is the $(q-1)$-dimensional unit simplex

$$S^{q-1} = \Big\{ t \in [\,0\,,1\,]^q : \sum_{i=1}^{q} t_i = 1 \Big\}; \tag{1.1a}$$

in component amount models, the experimental domain is the q-dimensional simplex

$$\mathcal{S} = \Big\{ t \in [\,0\,,1\,]^q : \sum_{i=1}^{q} t_i \leq 1 \Big\}. \tag{1.1b}$$

Under experimental conditions t, the response Y_t is taken to be a scalar random variable. Replications under identical conditions, or responses from distinct conditions are assumed to be uncorrelated with equal (unknown) variance σ^2. We follow Becker (1968), see also Cornell and Gorman (1978), in considering, for classical mixture experiments, an additive and homogeneous response. That is, we start from ν permutationally invariant and continuous functions f_j on the j-dimensional simplex, all of which are homogeneous of degree one, (here, $1 \leq \nu \leq q$ is a fixed integer). Define the k-dimensional regression vector $f, k = \sum_{j=1}^{\nu} \binom{q}{j}$, by

$$f(t) = (f_1(t_1), \dots, f_1(t_q), f_2(t_1, t_2), \dots, f_2(t_{q-1}, t_q), \dots,$$
$$f_\nu(t_1, \dots, t_\nu), \dots, f_\nu(t_{q-\nu+1}, \dots, t_q))', \tag{1.2}$$

where $t = (t_1, \ldots, t_q) \in \mathcal{S}$. We thus have

$$f(\alpha\, t) = \alpha\, f(t) \qquad \text{for all } 0 \leq \alpha \leq 1 \quad \text{and all } t \in \mathcal{S}\,. \tag{1.3a}$$

For avoiding trivialities we assume that the regression functions $f_1, \ldots, f_\nu$ are linearly independent over S^{q-1}. Note that by homogeneity of f this particularly implies $f_1(1) \neq 0$. Consequently, with c being the k-dimensional vector whose first q components equal $1/f_1(1)$ while all other components equal zero, we get

$$c' f(t) = \sum_{i=1}^{q} t_i \qquad \text{for all } t \in \mathcal{S}\,. \tag{1.3b}$$

For classical mixture experiments, we consider the linear regression model $\mathcal{M}$ for the expected response

$$\mathcal{M}\ :\ \mathrm{E}[\,Y_t\,] \,=\, \theta' f(t)\,, \qquad t \in S^{q-1}\,, \tag{1.4a}$$

where, as usual, θ is viewed as an unknown k-dimensional mean parameter vector. The corresponding component amount models $\widetilde{\mathcal{M}}$ is obtained by including the constant 1 into the regression model, simultaneously enlarging the experimental domain from S^{q-1} to $\mathcal{S}$,

$$\widetilde{\mathcal{M}}\ :\ \mathrm{E}[\,Y_t\,] = \vartheta_0 + \theta' f(t) = \widetilde{\theta}' \widetilde{f}(t), \text{ say}, \qquad t \in \mathcal{S}\,; \tag{1.4b}$$

(henceforth, a tilde in our notation refers to a mixture amount setting related to a mixture model via (1.4b)). Becker (1968) proposed three particular homogeneous mixture models $\mathcal{H}_1$, $\mathcal{H}_2$, $\mathcal{H}_3$ with (1.3a-b),

$$\begin{aligned}
\mathcal{H}_1\ :\ E[Y_t] &= \sum_{i=1}^{q} \vartheta_i t_i + \sum_{1 \leq i < j \leq q} \vartheta_{ij} \min\{t_i, t_j\} + \cdots \\
&\qquad + \sum_{1 \leq i_1 < \cdots < i_\nu \leq q} \vartheta_{i_1 \cdots i_\nu} \min\{t_{i_1}, \ldots, t_{i_\nu}\}, \\
\mathcal{H}_2\ :\ E[Y_t] &= \sum_{i=1}^{q} \vartheta_i\, t_i + \sum_{1 \leq i < j \leq q} \vartheta_{ij}\, \frac{t_i t_j}{t_i + t_j} + \cdots \\
&\qquad + \sum_{1 \leq i_1 < \cdots < i_\nu \leq q} \vartheta_{i_1 \cdots i_\nu} \frac{t_{i_1} \cdots t_{i_\nu}}{(t_{i_1} + \cdots + t_{i_\nu})^{\nu - 1}}, \\
\mathcal{H}_3\ :\ E[Y_t] &= \sum_{i=1}^{q} \vartheta_i\, t_i + \sum_{1 \leq i < j \leq q} \vartheta_{ij}\, (t_i t_j)^{1/2} + \cdots \\
&\qquad + \sum_{1 \leq i_1 < \cdots < i_\nu \leq q} \vartheta_{i_1 \cdots i_\nu}\, (t_{i_1} \cdots\, t_{i_\nu})^{1/\nu},
\end{aligned}$$

where $t \in S^{q-1}$; if in model $\mathcal{H}_2$ any denominator is zero, the value of the corresponding term is taken to be zero. For discussions on these models see for instance Snee (1973) and Becker (1978). We also refer to Draper and Pukelsheim (1998a, b) for detailed investigations of homogeneous mixture polynomials.

An experimental design ξ is a probability measure (on the experimental domain) having a finite number of support points. If ξ assigns weights $w_1, w_2, \ldots$ to its points of support, then the experimenter is directed to draw proportions $w_1, w_2, \ldots$ of all observations under the respective experimental conditions. With a mixture design ξ on S^{q-1} we associate its moment matrix, (in model (1.4a)),

$$M(\xi) = \int_{S^{q-1}} f(t) f'(t) \, \mathrm{d}\,\xi(t)\,; \tag{1.5a}$$

similarly, for a mixture amount design $\widetilde{\xi}$ on $\mathcal{S}$, its moment matrix in model (1.4b) is

$$\widetilde{M}(\widetilde{\xi}) \;=\; \int_{\mathcal{S}} \widetilde{f}(t) \widetilde{f}'(t) \, \mathrm{d}\,\widetilde{\xi}(t)\,. \tag{1.5b}$$

Of course, any mixture design ξ on S^{q-1} may be viewed as a mixture amount design. Then, the moment matrix $M(\xi)$ is a principal submatrix of $\widetilde{M}(\xi)$,

$$\widetilde{M}(\xi) = \left(\begin{array}{c|c} 1 & m(\xi)' \\ \hline m(\xi) & M(\xi) \end{array} \right)\,, \quad \text{with} \quad m(\xi) = \int_{S^{q-1}} f(t) \, \mathrm{d}\,\xi(t)\,. \tag{1.6}$$

Let c be the k-dimensional vector from (1.3b), and define the $k \times (k+1)$ matrix $C = (c \,|\, I_k)$, with I_k being the identity in $\mathbb{R}^{k \times k}$. Then $c' f(t) \equiv 1$ on S^{q-1}, hence $M(\xi)\, c = m(\xi)$ and $c' M(\xi)\, c = 1$, and (1.6) rewrites to

$$\widetilde{M}(\xi) \;=\; C' M(\xi)\, C \qquad \text{for all mixture designs } \xi \text{ on } S^{q-1}\,. \tag{1.7}$$

Homogeneity of the regression functions f and the relations (1.6) and (1.7) between the moment matrices are the fundamentals for our analysis:

In Section 2 we show that for many optimality criteria (namely, for all Loewner monotonic criteria), optimal mixture amount designs are supported by the origin and by S^{q-1}, only. Moreover, for specific criteria (among those are D-, A- and V-optimality), Proposition 2 establishes a simple method for constructing optimal mixture amount designs from optimal mixture ones (and vice versa): We only have to adjust the weight to be assigned to the origin, while − up to normalization − the weights assigned to support points from S^{q-1} remain unchanged.

The results unify and extend some of the known results on optimal mixture and mixture amount designs, as illustrated in Section 3. We comment on these results, and end up with some examples and counterexamples, demonstrating the power and limitations of the results.

2. An Admissibility Result

Design optimality usually aims at maximizing the moment matrices of designs in the sense of a statistically meaningful optimality criterion φ. Such a criterion is a $\mathbb{R}$-valued function, defined on the set of competing moment matrices, being isotonic w.r.t. the Loewner partial ordering, i.e., $A \geq_L B$ implies $\varphi(A) \geq \varphi(B)$; (the Loewner partial ordering is defined by $A \geq_L B$ iff $A - B$ is nonnegative definite). Mostly, determining φ-optimal designs is a difficult task, even when the optimization is done numerically. Therefore the question arises whether there are possibilities to restrict the search for a maximizing ξ to subclasses of designs.

One suitable subclass in this sense is formed by the admissible designs, where admissibility of a design ξ means that its moment matrix cannot be properly improved upon (w.r.t. the Loewner partial ordering) by the moment matrix of another design. That is, ξ is admissible iff for any other design τ, $M(\tau) \geq_L M(\xi)$ implies $M(\tau) = M(\xi)$, cf. e.g., Ehrenfeld (1956). Note that admissibility of a design depends on the underlying regression model.

In the mixture and component amount models (1.4a, b), continuity of the respective regression vectors and compactness of the experimental domains entail compactness of the associated sets of moment matrices of designs, and thereby it is not hard to see that in both models admissible designs exist, and, moreover, any design can be improved upon by an admissible one, (see Heiligers (1991, Lemma 1)). Actually, there is a close relation between the admissible mixture, and the admissible mixture amount designs.

Given $t \in \mathcal{S}$, we denote by δ_t the one-point design which assigns its mass to t only.

Proposition 1. *Let the regression functions f be homogeneous of degree one over $\mathcal{S}$. The set*

$$\widetilde{\mathcal{C}} = \{\, \widetilde{\xi} : \widetilde{\xi} \text{ is a design on } \mathcal{S} \text{ with } \operatorname{supp}(\widetilde{\xi}) \subset \{0\} \cup S^{q-1} \,\} \tag{2.1}$$

is a complete class of mixture amount designs, i.e., the moment matrix of any mixture amount design not in $\widetilde{\mathcal{C}}$ can properly be improved upon by that of a design from $\widetilde{\mathcal{C}}$. Moreover, under model (1.4b) *the mixture amount design $\widetilde{\xi}$ on $\mathcal{S}$ is admissible iff it decomposes into*

$$\widetilde{\xi} = \alpha\delta_0 + (1-\alpha)\,\xi \tag{2.2}$$

with some $0 \leq \alpha \leq 1$ and some admissible mixture design ξ on S^{q-1}, (here, admissibility of ξ refers to model (1.4a)*).*

Proof. (a) We start with verifying completeness of the class $\widetilde{\mathcal{C}}$ from (2.1). To this end, let $s \in \mathcal{S}$ with $s \neq 0$ and $s \notin S^{q-1}$. It suffices to show that the corresponding one-point design δ_s can properly be improved upon by a mixture amount design from $\widetilde{\mathcal{C}}$.

Let $\alpha = 1-\sum_{i=1}^{q} s_i$, thus $0 < \alpha < 1$, and define $t = (1-\alpha)^{-1}\, s \in S^{q-1}$. Consider the two-point design $\widetilde{\xi} \in \widetilde{\mathcal{C}}$ given by $\widetilde{\xi} = \alpha\,\delta_0 + (1-\alpha)\,\delta_t$. Observing that $f(0) = 0$ and $f(s) = (1-\alpha)\, f(t) \neq 0$, see (1.3a, b), we find

$$\begin{aligned}
\widetilde{M}(\widetilde{\xi}) - \widetilde{M}(\delta_s) &= \alpha\, \widetilde{f}(0)\widetilde{f}'(0) + (1-\alpha)\, \widetilde{f}(t)\widetilde{f}'(t) - \widetilde{f}(s)\widetilde{f}'(s) \\
&= \alpha \left(\begin{array}{c|c} 1 & 0_{1\times k} \\ \hline 0_{k\times 1} & 0_{k\times k}\end{array}\right) + (1-\alpha) \left(\begin{array}{c|c} 1 & f'(t) \\ \hline f(t) & f(t)f'(t)\end{array}\right) - \left(\begin{array}{c|c} 1 & f'(s) \\ \hline f(s) & f(s)f'(s)\end{array}\right) \\
&= (\alpha-\alpha^2) \left(\begin{array}{c|c} 0 & 0_{1\times k} \\ \hline 0_{k\times 1} & f(t)f'(t)\end{array}\right) = D, \quad \text{say;}
\end{aligned}$$

here, $0_{k\times 1}$ and $0_{k\times k}$ denote the zero vector and zero matrix in $\mathbb{R}^k$ and $\mathbb{R}^{k\times k}$, respectively. Obviously, D is nonnegative definite. Moreover, since $0 < \alpha < 1$ and $f(t) \neq 0$, we conclude $D \neq 0$, and $\widetilde{\xi}$ properly improves upon δ_s.

(b) We show that the one-point design δ_0 in 0 is admissible under the component amount model (1.4b). For, let $\widetilde{\xi}$ be a mixture amount design with $\widetilde{M}(\widetilde{\xi}) \geq_L \widetilde{M}(\delta_0)$. Since the regression vector $\widetilde{f} = (1\,, f')'$ contains the constant term 1, Loewner comparability of the moment matrices ensures that

$$0_{k\times 1} = \int_{\mathcal{S}} f(t)\, \mathrm{d}\,\widetilde{\delta}_0(t) = \int_{\mathcal{S}} f(t)\, \mathrm{d}\,\widetilde{\xi}(t), \tag{2.3}$$

see Theorem 2 in Heiligers (1991), see also Lemma 3.3 in Gaffke and Heiligers (1996a). Multiplying (2.3) from the left by c', with c from (1.3b), gives

$$0 = \sum_{t \in \mathrm{supp}\,(\widetilde{\xi})} \widetilde{\xi}(t)\, c' f(t) = \sum_{t \in \mathrm{supp}\,(\widetilde{\xi})} \Big(\widetilde{\xi}(t) \sum_{i=1}^{q} t_i\Big).$$

Because of $\sum_{i=1}^{q} t_i \geq 0$ for all $t = (t_1, \ldots, t_q)' \in \mathrm{supp}\,(\widetilde{\xi})$, with equality only if $t = 0_{q\times 1}$, it follows that $\widetilde{\xi}$ is the one-point measure in 0, and δ_0 is admissible.

(c) Let $\widetilde{\xi} \neq \delta_0$ be an admissible mixture amount design, thus $\widetilde{\xi}$ is of the form (2.2) with some $0 \leq \alpha < 1$ and some mixture design ξ. We show admissibility of ξ under the mixture model (1.4a).

Consider a design τ on S^{q-1} with $M(\tau) \geq_L M(\xi)$, and define the mixture amount design $\widetilde{\tau} = \alpha\,\delta_0 + (1-\alpha)\,\tau$. From (1.7), applied to τ, we get $\widetilde{M}(\widetilde{\tau}) = \alpha\widetilde{M}(\delta_0) + (1-\alpha)\, C' M(\tau)\, C$, and similarly, again applying (1.7), $\widetilde{M}(\widetilde{\xi}) = \alpha\widetilde{M}(\delta_0) + (1-\alpha) C' M(\xi)\, C$. Consequently, the matrix $\widetilde{D} = \widetilde{M}(\widetilde{\tau}) - \widetilde{M}(\widetilde{\xi}) = (1-\alpha) C'(M(\tau) - M(\xi))C$ is nonnegative definite, and therefore, due to admissibility of $\widetilde{\xi}$, it equals zero. Since $M(\tau) - M(\xi)$ is a diagonal submatrix of $\widetilde{D}$, it follows that $M(\tau) = M(\xi)$, ensuring admissibility of ξ.

(d) Let ξ be an admissible mixture design on S^{q-1}, and let $0 \le \alpha \le 1$. We prove admissibility (in model (1.4b)) of $\widetilde{\xi} = \alpha\delta_0 + (1-\alpha)\,\xi$.

In virtue of part (b) it suffices to consider the case of $\alpha < 1$. Let $\widetilde{\tau}$ be an admissible mixture amount design with $\widetilde{M}(\widetilde{\tau}) \ge_L \widetilde{M}(\widetilde{\xi})$, (such a design $\widetilde{\tau}$ exists, see Lemma 1 in Heiligers (1991)). By parts (a) (and (c)), $\widetilde{\tau} = \beta\delta_0 + (1-\beta)\,\tau$ with some $0 \le \beta \le 1$ and some (admissible) mixture design τ on S^{q-1}. From Theorem 2 in Heiligers (1991) we conclude

$$\begin{aligned} \int_{\mathcal{S}} f(t)\,\mathrm{d}\,\widetilde{\tau}(t) \qquad & \Big(= (1-\beta) m(\tau)\Big) \\ = \int_{\mathcal{S}} f(t)\,\mathrm{d}\,\widetilde{\xi}(t) \qquad & \Big(= (1-\alpha) m(\xi)\Big), \end{aligned} \tag{2.4}$$

$$\begin{aligned} \int_{\mathcal{S}} f(t)f'(t)\,\mathrm{d}\,\widetilde{\tau}(t) \qquad & \Big(= (1-\beta)\,M(\tau)\Big) \\ \ge_L \int_{\mathcal{S}} f(t)f'(t)\,\mathrm{d}\,\widetilde{\xi}(t) \qquad & \Big(= (1-\alpha) M(\xi)\Big). \end{aligned} \tag{2.5}$$

Let the vector c and the matrix C be as before. Multiplication of (2.4) by c' from the left directly yields $(1-\beta) = (1-\alpha)$. From (2.5) we hence get $M(\tau) \ge_L M(\xi)$, and therefore, observing admissibility of ξ, $M(\tau) = M(\xi)$. Since $\widetilde{\tau}$ is admissible, the identity $\widetilde{M}(\widetilde{\tau}) = \beta\widetilde{M}(\delta_0) + (1-\beta)\,C'M(\tau)C = \alpha\widetilde{M}(\delta_0) + (1-\alpha)\,C'M(\xi)C = \widetilde{M}(\widetilde{\xi})$, cf. (1.7), now entails admissibility of $\widetilde{\xi}$.

By obvious modifications in the proof to Proposition 1 it is seen that the assertions remain valid if the regression vector f is not homogeneous of degree one, but of degree $p \ge 1$, i.e., $f(\alpha\, t) = \alpha^p f(t)$ for all $0 \le \alpha \le 1$ and $t \in \mathcal{S}$. Hence, for example, in the quadratic Scheffé (and Kronecker) settings, the results from Draper, Heiligers and Pukelsheim (1999, 2000) ensure that the permutationally invariant and admissible mixture amount designs are supported by 0 and by barycenters of S^{q-1}, only; see also Theorem 4.4 of the latter paper for a smaller essentially complete class of admissible designs.

Usually, design admissibility does not imply any statistically meaningful optimality property, (see, however, Pukelsheim (1993), Chapter 10, for some optimality properties), but may be viewed only as a fundamental property, outruling the most inefficient designs. That is because admissibility depends on the design support only, but not on the associated weights, see e.g., Karlin and Studden (1966), Theorem 7.2. Moreover, although by Proposition 1 the reasonable mixture amount designs have the origin as the only possible support point outside the unit simplex S^{q-1}, this in general does not mean or imply that optimal mixture amount designs are directly obtainable from optimal mixture designs by adjusting the weight α assigned to 0 only. Nevertheless, Proposition 1 allows to derive simple transformation rules for the important and popular D-, A-, and V-criteria, (the latter is also called I-criterion), which aim at minimizing the respective generalized variance, average variance of parameter estimates, and average variance of the predicted regression function over the experimental region, see e.g., Atkinson and Donev (1992, Chapter 10), or Pukelsheim (1993, Chapter 6), see also Studden (1977).

Proposition 2. *Suppose that the regression functions f from* (1.4b) *are homogeneous of degree one over $\mathcal{S}$. Then the mixture amount design $\widetilde{\xi}^*$ in model* (1.4b) *is D-optimal (A-optimal, V-optimal) iff $\widetilde{\xi}^* = \alpha^* \delta_0 + (1-\alpha^*)\,\xi^*$, where ξ^* is a D-optimal (A-optimal, V-optimal) mixture design in model* (1.4a), *and $\alpha^* = (1+\sqrt{\beta^*})^{-1}$ with*

$$\beta^* = \begin{cases} k^2\,, & \textit{for D-optimality}, \\ \frac{1}{1+c'c}\,\mathrm{trace}[\,M^{-1}(\xi_A)\,]\,, & \textit{for A-optimality}, \ (\xi_A = \xi^*), \\ \frac{(q+1)!}{2}\displaystyle\int_{S^{q-1}} f'(u)M^{-1}(\xi_V)f(u)\mathrm{d}u_1\cdots\mathrm{d}u_{q-1}, & \textit{for V-optimality}, \ (\xi_V = \xi^*). \end{cases}$$

Proof. Consider a mixture amount design $\widetilde{\xi} = \alpha\,\delta_0 + (1-\alpha)\,\xi$ from the complete class $\widetilde{\mathcal{C}}$ (2.1). Abbreviating $\widetilde{M} = \widetilde{M}(\widetilde{\xi})$, $M = M(\xi)$, and $m = m(\xi)$, we find

$$\widetilde{M} = \left(\begin{array}{c|c} 1 & (1-\alpha)\,m' \\ \hline (1-\alpha)\,m & (1-\alpha)\,M \end{array}\right)\,,$$

see (1.7). Recall that $m = M\,c$, hence $m'M^+m = c'Mc = 1$, where M^+ is the Moore-Penrose inverse of M. We hence compute

$$\det[\widetilde{M}] = (1-(1-\alpha)m'M^+m)\det[(1-\alpha)M] = \alpha\,(1-\alpha)^k\det[M]\,, \tag{2.6}$$

see e.g., Theorem 13.3.8 and Section 14.8 in Harville (1997). It particularly follows that $\widetilde{M}$ is nonsingular iff M is nonsingular and $0 < \alpha < 1$. Moreover, if $\widetilde{M}$ is nonsingular, and consequently $c = M^{-1}m$, then

$$\widetilde{M}^{-1} = \frac{1}{\alpha}\left(\begin{array}{c|c} 1 & -c' \\ \hline -c & cc' + \frac{\alpha}{1-\alpha}M^{-1} \end{array}\right)\,,$$

and, obviously,

$$\mathrm{trace}[\widetilde{M}^{-1}] = \frac{1+c'c}{\alpha} + \frac{1}{1-\alpha}\,\mathrm{trace}[M^{-1}]. \tag{2.7}$$

Finally, for the $(k+1)\times(k+1)$ matrix $\widetilde{F} = \int_{\mathcal{S}} \tilde{f}(s)\tilde{f}'(s)\,\mathrm{d}s$, homogeneity of the regression vector f implies

$$\widetilde{F} = \int_{S^{q-1}} \left(\begin{array}{c|c} \frac{1}{q} & \frac{1}{q+1}\,f'(u) \\ \hline \frac{1}{q+1}\,f(u) & \frac{1}{q+2}\,f(u)f'(u) \end{array}\right)\,\mathrm{d}u_1\cdots\mathrm{d}u_{q-1}\,.$$

Utilizing the formula $\int_{S^{q-1}} 1 \mathrm{d}u_1 \cdots \mathrm{d}u_{q-1} = 1/(q{-}1)!$, and observing that $c' f(u) \equiv 1$ on S^{q-1}, we therefore compute

$$\int_{\mathcal{S}} \tilde{f}'(s) \widetilde{M}^{-1} \tilde{f}(s) \, \mathrm{d}\, s = \mathrm{trace}[\widetilde{M}^{-1} \widetilde{F}]$$
$$= \frac{2}{\alpha\,(q{+}2)!} + \frac{1}{(1{-}\alpha)\,(q{+}2)} \int_{S^{q-1}} f'(u) M^{-1} f(u) \, \mathrm{d}u_1 \cdots \mathrm{d}u_{q-1} \, . \qquad (2.8)$$

Since the *D*-, *A*-, and *V*-criterion are strictly monotonic w.r.t. the Loewner partial ordering, the optimal mixture amount designs are admissible, and thus, by Proposition 1, they belong to the complete class $\widetilde{\mathcal{C}}$. The assertions now follow from (2.6), (2.7) and (2.8) by straightforward computations.

A more stringent version of Proposition 2 carries over to linear optimality for the linear parameter function $\widetilde{A}\widetilde{\Theta}$, with some fixed $\widetilde{A} = (a \,|\, A) \in I\!R^{r \times (k+1)}$; (as special cases, these criteria lead to *A*-, and *V*-optimality, when $\widetilde{A} = I_{k+1}$ and $\widetilde{A} = \widetilde{F}$, respectively, and to scalar optimality, when $r = 1$). Then, arguing as in the proof to Proposition 2, we obtain for $\tilde{\xi} = \alpha\, \delta_0 + (1{-}\alpha)\, \xi \in \widetilde{\mathcal{C}}$, $\mathrm{trace}[\widetilde{L}\widetilde{M}^{-1}] = (1/\alpha)(l_{11} - 2 l_1' c + c' L c) + [1/(1-\alpha)] \mathrm{trace}[L M^{-1}]$. with $\widetilde{L}, l_{11}, l_1$ and L defined by

$$\widetilde{L} = \left(\begin{array}{c|c} l_{11} & l_1' \\ \hline l_1 & L \end{array} \right) = \widetilde{A}' \widetilde{A}.$$

Note however that, depending on $\widetilde{A}$, a linear optimal design need not even belong to the complete class $\widetilde{\mathcal{C}}$, but of course, among the optimal designs, there is an admissible one.

It follows, that $\tilde{\xi} = \alpha\, \delta_0 + (1{-}\alpha)\, \xi \in \widetilde{\mathcal{C}}$ is linear optimal for $\widetilde{A}\widetilde{\Theta}$ (among all mixture amount designs) iff ξ is linear optimal for $A\Theta$ and $\alpha = (1{+}\sqrt{\beta})^{-1}$, with $\beta = (\mathrm{trace}[A\, M^{-1}\, A'] / (a'a - 2a'Ac + c'A'Ac)^{-1}$. Consequently, given a linear optimal mixture design ξ for $A\Theta$, an optimal mitxure amount design for $(a \,|\, A)\widetilde{\Theta}$ is directly obtainable. Conversely, for obtaining a linear optimal mixture design for $A\Theta$ from an optimal mixture amount design $\tilde{\xi}$ for $(a \,|\, A)\widetilde{\Theta}$ we firstly have to find an optimal design in the complete class $\widetilde{\mathcal{C}}$. This one can be constructed by the method utilized in the proof to part (a) of Proposition 1; an explicit description, however, is somewhat technical in details, and we therefore omit it.

3. Examples and Discussions

Of course, an application of Proposition 2 for determining *D*-, *A*- or *V*-optimal designs on $\mathcal{S}$ (resp. on S^{q-1}) requires knowledge of the optimal ones on S^{q-1} (resp. $\mathcal{S}$). In most setups, the optimal mixture or mixture amount designs have to be computed numerically. Regarding this problem we found in our numerical examples that the Quasi-Newton method proposed by Gaffke and

Heiligers (1996a), see also Gaffke and Heiligers (1996b), is remarkably stable, and shows an excellent convergence behavior. We omit the details; instead we will focus here on examples where explicit results on optimal designs are available.

Becker models

For the quadratic ($\nu = 2$) Becker models $\mathcal{H}_1$, $\mathcal{H}_2$, $\mathcal{H}_3$ from Section 1 in two variables ($q = 2$), the D-optimal designs are given in Hilgers (1991, p.200), see also Liu and Neudecker (1997, Theorem 3), and for Becker's minimum model $\mathcal{H}_1$ with $q = \nu \geq 2$ in Hilgers (2000, Theorem 2.1). From these results and our Proposition 2 we directly obtain, (see also Hilgers (1991, p.197), and Hilgers (1999, Lemma 2.1)),

Corollary 3.
(a) *For model $\widetilde{\mathcal{H}}_1$ with $\nu = q \geq 2$, the D-optimal mixture amount design assigns equal mass $\alpha_D = 1/2^q$ to 0 and to all barycenters of S^{q-1} up to depth q.*
(b) *For models $\widetilde{\mathcal{H}}_2$ and $\widetilde{\mathcal{H}}_3$ with $\nu = q = 2$, the D-optimal mixture amount design assigns equal mass $\alpha_D = 1/4$ to $(0\,,0)$, $(1\,,0)$, $(0\,,1)$ and $(1/2\,,1/2)$.*

In the minimum model $\widetilde{\mathcal{H}}_1$ with $\nu < q$ it is known that the D-optimal mixture amount design does not spread its mass uniformly to 0 and to the barycenters of S^{q-1} of depth up to ν, cf. Hilgers (2000, Theorem 2.1). However, from Lemma 2.1 in that paper, combined with our Proposition 2, it follows that the optimal design assigns its mass to 0 and to the barycenters of S^{q-1} up to depth q, and hence the D-optimal design problem becomes an allocation problem. Numerical results, for $2 \leq \nu < q \leq 5$, are easily derived from Table 1 in Hilgers (2000).

Liu and Neudecker (1997) also obtain A- and V-optimal allocation designs on S^{q-1} for models $\mathcal{H}_1$, $\mathcal{H}_2$, $\mathcal{H}_3$, i.e., they solve the design problems within the specific subclass of designs being supported by the barycenters of S^{q-1} up to depth ν. Actually, the general equivalence theory for optimal designs, see e.g., Pukelsheim (1993, Section 7), ensures that for the minimum model $\mathcal{H}_1$ the A- and V-optimal designs are concentrated on the barycenters of S^{q-1} only, (combine the reasoning as in Atwood (1969, Theorem 2.1), with symmetry arguments as in Farrell, Kiefer and Walbran (1967, p.119), for example). Hence, the optimal allocation design for model $\mathcal{H}_1$ with $\nu = q$ from Theorem 1 and Theorem 4 in Liu and Neudecker (1997) are in fact optimal among all designs.

Corollary 4. *For model $\widetilde{\mathcal{H}}_1$ with $\nu = q \geq 2$,*
(a) *the A-optimal mixture amount design assigns mass $\alpha_A = (\sqrt{1+q} + q(1+\sqrt{2})^{q-1})^{-1}$ to 0, and mass $\alpha_A \cdot \sqrt{d^2\, 2^{q-d}/(1+q)}$ to each barycenter of S^{q-1} of depth d, $1 \leq d \leq q$;*
(b) *the V-optimal mixture amount design assigns mass $\alpha_V = (1 + \sum_{d=1}^{q} \sqrt{\binom{q}{d}})^{-1}$ to 0, and mass $\alpha_V \cdot \sqrt{1/\binom{q}{d}}$ to each barycenter of S^{q-1} of depth d, $1 \leq d \leq q$.*

Proof. The assertions follow by some lengthy computations from Proposition 2, combined with Theorems 1 and 4 in Liu and Neudecker (1997), or directly from Corollary 1 in Pukelsheim and Torsney (1991), observing that the collocation matrix built in the $\tilde{f}$ vector evaluated at 0 and all barycenters of S^{q-1} is a principal block-triangular matrix, whose inverse can be explicitly determined, (see the appendix in Liu and Neudecker (1997)).

As one might have expected, the optimal weight assigned to 0 monotonously tends to zero with an increasing number q of mixture ingredients; Figure 1 displays the weights for some values of q. Thus, for large q, the optimal mixture amount designs mimic their respective mixture counterparts. This, however, is not true for all setups and all criteria, (see the E-optimal design for the 1-tic model in Hilgers and Bauer (1995, p.245)).

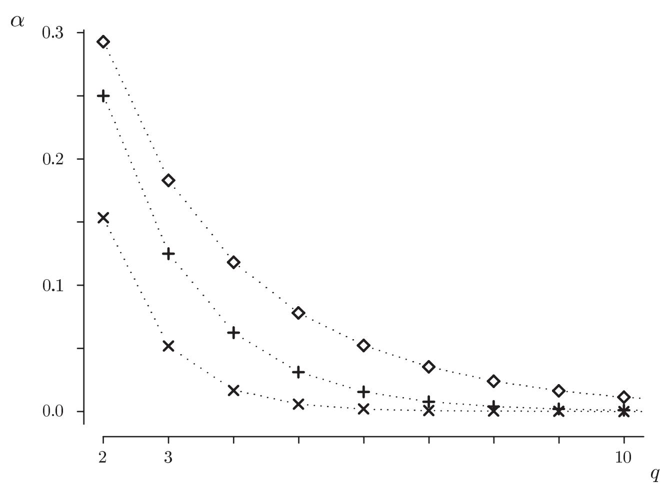

Figure 1. Weight α assigned to 0 by an optimal mixture amount design for the minimum model $\widetilde{\mathcal{H}}_1$ with $q = 2, \ldots, 10$. The crosses, stars, and diamonds give the respective D-, A-, and V-optimal weights from Corollaries 3 and 4.

Scheffé's ν-tic model

The regression vector f in the ν-tic model with $\nu \geq 2$,

$$\mathcal{S}_\nu \; : \; E[\,Y_t\,] = \sum_{\ell=1}^{\nu} \sum_{1 \leq i_1 < \cdots < i_\ell \leq q} \vartheta_{i_1 \cdots i_\ell} \prod_{j=1}^{\ell} t_{i_j} \, , \qquad t \in S^{q-1},$$

does obviously not satisfy our basic homogenity assumptions (1.3a), and hence the Transformation Proposition 2 is not directly applicable for obtaining optimal ν-tic amount designs. The arguments given in the proof to this proposition, however, ensure that the assertions remain valid if the following conditions hold:

$$\text{The optimal mixture amount design is concentrated on } 0 \cup S^{q-1}. \tag{3.1a}$$

$$\begin{array}{l}\text{The regression vector } f \text{(in the mixture model) satisfies} \\ \text{(i) } f(0) = 0, \text{ and (ii) } c'f \equiv 1 \text{ on } S^{q-1} \text{ for some vector } c.\end{array} \tag{3.1b}$$

Note that for D-optimality, part (ii) of condition (3.1b) may be replaced by the assumption $m'Mm = 1$ for all mixture designs ξ, see formulae (2.6) in the proof to Proposition 2.

Both conditions are satisfied in the ν-tic model: For, validity of (3.1b) is obvious; property (3.1a) follows from Lemma 2.1 in Hilgers and Bauer (1995) for the D-criterion. Indeed, the arguments given there carry over to the A- and V-criterion, (and, more gernerally, to any strictly Loewner isotonic optimality criterion).

Corollary 5.

(a) *For the ν-tic component amount model with $\nu = 2, \nu = 3$ or $\nu = q$, the D-optimal design assigns equal mass to the origin* 0 *and to all barycenters of S^{q-1} of depth up to ν.*

(b) *For the q-tic component amount model ($\nu = q$), the A-optimal design assigns mass $\alpha_A = (1+q\,2^{q/2}(1+\sqrt{2})^{q-1})^{-1}$ to the origin* 0*, and masses $\alpha_A \cdot d\,2^{(q-d)/2}$ to each barycenter of S^{q-1} of depth d, $1 \leq d \leq q$.*

Proof. We outline the proof. The statements on D-optimality follow immediately from our (modified) Proposition 2, combined with the known results on optimal mixture design, given in Atwood (1969), Theorem 2.4, Kiefer (1961), Section 5 and Uranisi (1964), Theorem 2. Regarding A-optimality, we remark that the optimal mixture design (in the q-tic model) is saturated, and hence it can be constructed by applying Corollary 1 in Pukelsheim and Torsney (1991), (see also the appendix in Liu and Neudecker (1995) for the inverse of the collocation matrix). The A-optimal mixture amount design is then obtained from Proposition 2.

It should be noted that part (a) of Corollary 5 restates partially Theorem 2.1 in Hilgers and Bauer (1995). We also note that the V-optimal design in the q-tic component amount model can be obtained from Theorem 2 in Liu and Neudecker (1995); the description, however, is somewhat technical, and is therefore omitted.

Counterexamples

From the following three examples it transpires that in general neither (3.1a) nor (3.1b) can be omitted without affecting validity of Proposition 2. Our first example refers to a model satisfying the basic homogenity assumptions (1.3a) but violating (1.3b). Consider the mixture and mixture amount setups $f(t) = (\min\{t_1, t_2\}, \min\{t_1, t_3\}, \min\{t_2,\ t_3\})'\,, t \in S^{q-1}$ and $\widetilde{f}(t) = (1, \min\{t_1, t_2\}, \min\{t_1, t_3\},\ \min\{t_2, t_3\})'$, $t \in \mathcal{S}$. Note that the regression vector f is homogeneous of degree 1 and $f(0) = 0$, but no nontrivial linear transformation of f is constant over S^{q-1}. Using the famous Kiefer and Wolfowitz (1960) equivalence theorem, a little algebra shows that the design

$$\xi_D(x) = \begin{cases} \dfrac{8}{27}, \text{ if } x = (1/2\,, 1/2\,, 0) \quad \text{or a permutation thereof,} \\ \dfrac{3}{27}, \text{ if } x = (1/3\,, 1/3\,, 1/3)\,, \end{cases}$$

is the D-optimal design mixture design. However, the design $\widetilde{\xi}_0 = (1/4)\,\delta_0 + (3/4)\,\xi_D$ derived therefrom by applying Proposition 2 is not D-optimal in the corresponding mixture amount setup. For, here we have

$$(\widetilde{M}(\widetilde{\xi}_0))^{-1} = \frac{1}{131}\begin{pmatrix} 180 & -252 & -252 & -252 \\ -252 & 6012 & 1062 & 1062 \\ -252 & 1062 & 6012 & 1062 \\ -252 & 1062 & 1062 & 6012 \end{pmatrix},$$

hence, abbreviating $g(x) = \widetilde{f}'(x)(\widetilde{M}(\widetilde{\xi}_0))^{-1}\widetilde{f}(x)$, we find $\max_{x\in\mathcal{S}}\, g(x) \geq g(1/3, 1/3, 1/3) = 972/131 > 4$ and the equivalence theorem for D-optimality entails non-optimality of $\widetilde{\xi}_0$. In fact, it can be shown that

$$\widetilde{\xi}_D(x) = \begin{cases} \dfrac{1}{6}\,, \text{ if } x = (0\,, 0\,, 0)\,, \\ \dfrac{2}{9}\,, \text{ if } x = (1/2\,, 1/2\,, 0) \quad \text{or a permutation thereof}\,, \\ \dfrac{1}{6}\,, \text{ if } x = (1/3\,, 1/3\,, 1/3)\,. \end{cases}$$

is the D-optimal design in the corresponding mixture amount setup.

Next we demonstrate that in a setups satisfying (3.1a) but violating (3.1b), the optimal mixture amount design is not necessarily a convex combination of the one-point design in 0 and of the corresponding optimal mixture design. To this end, we consider the A-optimal design problem for the mixture and mixture amount setups $f(t) = (t_1t_2, t_1t_3, t_2t_3)'$, $t \in S^{q-1}$ and $\widetilde{f}(t) = (1, t_1t_2, t_1t_3, t_2t_3)'$, $t \in \mathcal{S}$. Note that the regression vector f is homogeneous of degree 2, and hence by Proposition 1 condition (3.1a) is fulfilled. Obviously $f(0) = 0$, but no nontrivial linear transformation of f is constant over S^{q-1}. Based on the equivalence theorem for A-optimality, cf. e.g., Kiefer (1974), a little algebra shows that the uniform design ξ_A on the barycenters of S^{q-1} of depth 2 is the optimal mixture design, (actually, ξ_A is uniformly optimal for all permutationally invariant and concave criteria). Moreover, utilizing Corollary 1 in Pukelsheim and Torsney (1991), it is not hard to see that the mixture amount design $\widetilde{\xi}_0 = (7/19)\,\delta_0 + (12/19)\,\xi_A$ is A-optimal among all convex combinations of δ_0 and ξ_A, and

$$(\widetilde{M}(\widetilde{\xi}_0))^{-2} = \frac{1}{7}\begin{pmatrix} 2527 & -15884 & -15884 & -15884 \\ -15884 & 127072 & 86640 & 86640 \\ -15884 & 86640 & 127072 & 86640 \\ -15884 & 86640 & 86640 & 127072 \end{pmatrix}.$$

Abbreviating $g(x) = \widetilde{f}'(x)(\widetilde{M}(\widetilde{\xi}_0))^{-2}\widetilde{f}(x)$, we find $\max_{x\in\mathcal{S}} g(x) \ge g(1/3,1/3,1/3)$ $= 82669/189 > 361 = \max_{x\in\mathrm{supp}\,(\widetilde{\xi}_0)} g(x)$, and the equivalence theorem for A-optimality entails non-optimality of $\widetilde{\xi}_0$. In fact, it can be shown here that the A-optimal design is a five-point design, supported by 0 and the barycenters of S^{q-1} of depth 2 and 3,

$$\widetilde{\xi}_A(x) \approx \begin{cases} 0.35307\,, \text{ if } x = (0\,,0\,,0)\,, \\ 0.19927\,, \text{ if } x = (1/2\,,1/2\,,0) \quad \text{or a permutation thereof}\,, \\ 0.04913\,, \text{ if } x = (1/3\,,1/3\,,1/3)\,. \end{cases}$$

Finally, we show that in non-homogeneous setups satisfying (3.1b), an optimal mixture amount design does not necessarily satisfy (3.1a). To this end, consider D-optimality in the quadratic Darroch and Waller (1985) mixture model in $q = 3$ variables,

$$f(t) = (t_1\,,t_2\,,t_3\,,t_1(1-t_1)\,,t_2(1-t_2)\,,t_3(1-t_3))', \qquad t \in S^{q-1}\,,$$

and in the corresponding component amount model,

$$\widetilde{f}(t) = (1\,,t_1\,,t_2\,,t_3\,,t_1(1-t_1)\,,t_2(1-t_2)\,,t_3(1-t_3))', \qquad t \in \mathcal{S}\,.$$

Obviously, property (3.1b) is met, taking $c = (1\,,1\,,1\,,0\,,0\,,0)'$ in part (ii). Note that the regression vector f is not homogeneous (of any degree $p \ge 1$), and hence Proposition 1 does not ensure that an optimal (admissible) design fulfills (3.1a). (Actually, from Lemma 1 in Heiligers (1991) and Corollary 1 in Heiligers (1992), see also Lemma 4.1 in Gaffke and Heiligers (1996), it follows that for any admissible and permutationally invariant mixture amount design the only possible support points are the origin 0, the barycenters of S^{q-1}, and multiples in $\mathcal{S}$ of these barycenters, (at most one in the relative interior of the line segments joining the origin with the individual barycenters)).

By Kiefer and Wolfowitz (1960), it is not hard to see that the D-optimal mixture design on S^{q-1} gives equal mass 1/6 to the barycenters of S^{q-1} of depth 1 and 2, (see also Kiefer (1961), and Zhang and Guan (1992)). Hence, if (3.1a) would be satisfied, then the uniform design $\widetilde{\xi}_0$ supported by 0 and the barycenters of S^{q-1} of depth 1 and 2 would be D-optimal. However, for $\widetilde{\xi}_0$ we compute

$$(\widetilde{M}(\widetilde{\xi}_0))^{-1} = \begin{pmatrix} 7 & -7 & -7 & -7 & 0 & 0 & 0 \\ -7 & 14 & 7 & 7 & -14 & 0 & 0 \\ -7 & 7 & 14 & 7 & 0 & -14 & 0 \\ -7 & 7 & 7 & 14 & 0 & 0 & -14 \\ 0 & -14 & 0 & 0 & 112 & -28 & -28 \\ 0 & 0 & -14 & 0 & -28 & 112 & -28 \\ 0 & 0 & 0 & -14 & -28 & -28 & 112 \end{pmatrix},$$

and, abbreviating $g(x) = \widetilde{f}'(x)(\widetilde{M}(\widetilde{\xi}_0))^{-1}\widetilde{f}(x)$, $\max_{x\in\mathcal{S}} g(x) \geq g(1/3\,,0\,,0) = 595/81 > \max_{x\in\mathrm{supp}\,(\widetilde{\xi}_0)} g(x) = 7$. Consequently, by the Kiefer and Wolfowitz Theorem, $\widetilde{\xi}_0$ is not D-optimal, and therefore (3.1a) is not fulfilled in this setup. Actually, by numerical computations, the D-optimal mixture amount design having guaranteed D-efficiency $\geq 1-10^{-7}$ for the Darroch-Waller model is found to be

$$\widetilde{\xi}_D(x) \approx \begin{cases} 0.11341\,, \text{ if } x = (0\,,0\,,0)\,, \\ 0.14268\,, \text{ if } x = (1\,,0\,,0) \quad \text{or a permuation thereof}\,, \\ 0.12584\,, \text{ if } x = (1/2\,,1/2\,,0) \quad \text{or a permutation thereof}\,, \\ 0.02701\,, \text{ if } x = (0.38245\,,0\,,0) \quad \text{or a permuation thereof}\,. \end{cases}$$

We note that for the Darroch-Waller model with $q \geq 4$, there is much numerical evidence that the D-optimal mixture amount design satisfies (3.1a), at least for $q \leq 20$. However, we do not have a theoretical explanation for this observation.

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