Efficient algorithms for constructing D- and I-optimal exact designs for linear and non-linear models in mixture experiments

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Supplementary Material A

The Gâteax derivative at p in the direction of $[h]$ is defined

$$G_\Phi(p,h) \quad = \quad \lim_{\beta\to 0^+} \frac{\Phi(p+\beta h)-\Phi(p)}{\beta} = \lim_{\beta\to 0^+} \frac{\Psi[M(\mathbb{P}+\beta\mathbb{H})]-\Psi[M(\mathbb{P})]}{\beta}.$$

Considering the first-order Taylor expansion serie of the non-linear functions of the elements of the information matrix

$$M(p+\beta h) \approx M(\mathbb{P})+\beta\sum_{i=1}^{q} h_i \frac{\partial M(\mathbb{P})}{\partial p_i},$$

it results

$$\begin{aligned} G_\Phi(p,h) &= \lim_{\beta\to 0^+} \frac{\Psi\left[M(\mathbb{P})+\beta\left(\sum_{i=1}^{q} h_i \frac{\partial M(\mathbb{P})}{\partial p_i}\right)\right]-\Psi[M(\mathbb{P})]}{\beta} \\ &= G_\Psi\left[M(\mathbb{P}),\sum_{i=1}^{q} h_i \frac{\partial M(\mathbb{P})}{\partial p_i}\right]. \end{aligned}$$

The directional derivative and the Gâteaux are related by

$$F_\Phi(p,h) \quad = \quad G_\Phi(p,h-p),$$

so that

$$F_\Phi(p,h) \quad = \quad F_\Psi\left[M(\mathbb{P}),M(\mathbb{P})+\sum_{i=1}^{q}(h_i-p_i)\frac{\partial M(\mathbb{P})}{\partial p_i}\right]$$

and, if $h=e_j$, then

$$F_\Phi(p,e_j) \quad = \quad F_\Psi\left[M(\mathbb{P}),M(\mathbb{P})+\frac{\partial M(\mathbb{P})}{\partial p_j}-\sum_{i=1}^{q} p_i \frac{\partial M(\mathbb{P})}{\partial p_i}\right].$$

For D-optimality criterion $\Psi[M(\mathbb{P})]=Ln|M(\mathbb{P})|$, then

$$\frac{\partial \Psi[M(\mathbb{P})]}{\partial p_r} = \sum_{i=1}^{q}\sum_{j=1}^{q} m_{ij}^{-1}(p)\frac{\partial m_{ij}(p)}{\partial p_r} = \mathrm{Tr}\left[M^{-1}(\mathbb{P})\frac{\partial M(\mathbb{P})}{\partial p_r}\right],$$

where $m_{ij}(p)$ is the $(i,j)^{th}$ entry of the matrix $M(\mathbb{P})$. So,

$$F_{\Phi_D}(p,e_j) = \mathrm{Tr}\left[M^{-1}(\mathbb{P})\frac{\partial M(\mathbb{P})}{\partial p_j}\right] - \sum_{i=1}^{q} p_i \mathrm{Tr}\left[M^{-1}(\mathbb{P})\frac{\partial M(\mathbb{P})}{\partial p_i}\right].$$

Supplementary Material B

In the case of unrestricted examples, the scenarios were:

1. **Random Designs (RD).** For each $p_i \in \xi_j,\ i = 1, \ldots, n,\ j = 1, \ldots, M$ generate $u_{ik} \sim U(0,1),\ k = 1, \ldots, q$, so that $p_i = (p_1^i, \ldots, p_q^i)$ being $p_k^i = \dfrac{u_{ik}}{\sum_{k=1}^{q} u_{ik}}$.

2. **Random Uniform Desings (RUD).** Some authors warn the previous sampling methodology can generate non-uniform distributions. Then, one way of obtaining a uniform sample from the simplex is $p_k^i = \dfrac{-ln(u_{ik})}{\sum_{k=1}^{q} -ln(u_{ik})}$, which generates IID random samples from an exponential distribution.

3. **Vertex-and-centroid-near-point Designs (VD)**. Let C be the matrix containing the vertices, the overall centroid and the centroid of all lower dimensional simplices of a $(q-1)$-dimensional simplex. These points were obtained invoking the program *crvtave* of Piepel (REF). From an uniform grid G,

$$G = \{p = (p_1, \ldots, p_q) \in \mathcal{S} \ / \ p_i = \frac{1}{k} \cdot j,\ j \in \mathbb{N},\ 0 \leq j \leq k,\ i = 1, \ldots, q\},$$

construct a distance matrix D, where $d_{ij} = \parallel p_i - p_j \parallel_2$, $p_i \in C$ and $p_j \in G$. Fill $M/2$ initial designs with a random sample with replacement of the $\dfrac{M \cdot N}{2}$ points with lower d_{ij} values. The $M/2$ remaining designs will be filled with random points of G.

When there are limitations over the ingredient quantities, previous scenarios could not be valid. So it is necessary to define new situations:

1. **Random Restricted Designs (RRD).**
   i) Calculate $dif_i = U_i - L_i$ for each $i = 1, \ldots, q$.
   ii) Sort the list of dif_i in increasing order: $dif_{i_1}, \ldots, dif_{i_q}$.
   iii) For each $p_i \in \xi_j$, $i = 1, \ldots, n,\ j = 1, \ldots, M$, generate $u_{ik} \sim U(0,1)$, $k = 1, \ldots, q-1$ satisfying $L_{i_j} \leq u_{ik} \leq U_{i_j}$, $j = 1, \ldots, q-1$, so that $p_i = (u_{i1}, \ldots, u_{i1(q-1)}, u_{iq})$ being $u_{iq} = 1 - \sum_{k=1}^{q-1} u_{ik}$.
   iv) If $L_{i_q} \leq u_{iq} \leq U_{i_q}$, then add p_i to the design. Otherwise, return to iii).

2. **Extreme Vertices Design (EVD).** Generate the vertices, midpoints of edges connecting these vertices and overall centroid of the constrained region and collect them into the matrix V. Create interior points by averaging all possible pairs of points of V (*Fillv* function of *mixexp* R package) and collect them into the matrix F. Fill each ξ_j, $j = 1, \ldots, M$ with a N-size random sample with replacement of F.

3. **Semi-Extreme Vertices Design (SEVD).** Designs are analogously constructed to previous scenario, except that V points are removed from F.

Figures 1 and 2 illustrate a sample of each class of the unrestricted and restricted designs previously defined. They were graphically depicted using the *mixexp* R package. In these graphs, simplex vertices represent pure components, edges correspond to binary blends and simplex interior points contain all three-component systems.

(*a*) (*b*) (*c*)

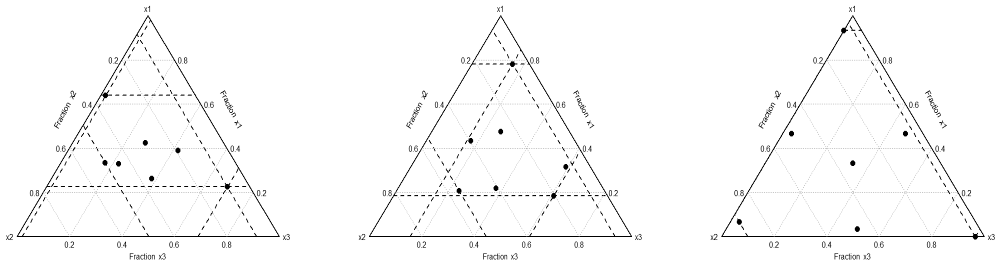

Figure 1: *Samples of Random Design (RD) (a), Random Uniform Designs (RUD) (b) and Vertex-and-centroid-near-point Design (VD) (c) with 7 design points.*

(*a*) (*b*) (*c*)

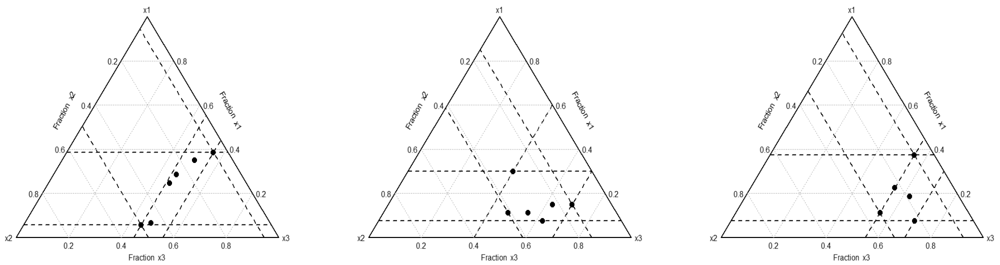

Figure 2: *Samples of Random Restricted Design (RRD) (a), Extreme Vertices Design (EVD) (b) and Semi-Extreme Vertices Design (SEVD) (d) with 6 design points.*

Supplementary material C

Table 1: *Relative D-efficiencies for model (7) under different scenarios (RD, RUD and VD).* $^{(*)}$ *indicates that the obtained result does not depend on a set of candidate points.*

No. ingredients	3			
No. points	Algorithm	RD	RUD	VD
7	GA	1	1	1
	KLA	0.6899	0.8148	1
	CEA$^{(*)}$	1	1	1
14	GA	1	1	1
	KLA	0.6865	0.8308	1
	CEA$^{(*)}$	1	1	1
18	GA	1	1	1
	KLA	0.7861	0.8772	0.9139
	MA$^{(*)}$	1	1	1
	CEA$^{(*)}$	1	1	1
No. ingredients	5			
No. points	Algorithm	RD	RUD	VD
25	GA	0.3433	0.8584	1
	KLA	0.2403	0.4334	1
	CEA$^{(*)}$	1	1	1
50	GA	0.5772	0.7611	1
	KLA	0.3002	0.5137	0.9978
	CEA$^{(*)}$	1	1	1

Table 2: *Relative I-efficiencies for second-order model under different scenarios (RD, RUD and VD).* $^{(*)}$ *indicates that the obtained result does not depend on a set of candidate points.*

No. ingredients	3			
No. points	Algorithm	RD	RUD	VD
6	GA	0.9842	0.9887	0.9882
	KLA	0.7463	0.9040	0.9881
	CEA$^{(*)}$	1	1	1
7	GA	0.9606	0.9605	0.9894
	KLA	0.8431	0.8814	0.9861
	CEA$^{(*)}$	0.9953	0.9953	0.9953
8	GA	0.9803	0.9918	0.9644
	KLA	0.8208	0.8943	0.9875
	CEA$^{(*)}$	1	1	1
18	GA	0.9719	0.9696	0.9997
	KLA	0.8154	0.9281	0.9978
	MA$^{(*)}$	1	1	1
	CEA$^{(*)}$	1	1	1
30	GA	0.9452	0.9945	0.9459
	KLA	0.9222	0.9293	0.9981
	CEA$^{(*)}$	1	1	1
No. ingredients	4			
No. points	Algorithm	RD	RUD	VD
15	GA	0.8611	0.9604	0.9801
	KLA	0.5839	0.7565	0.9543
	CEA$^{(*)}$	1	1	1
16	GA	0.7468	0.8777	1
	KLA	0.5452	0.7591	0.9994
	CEA$^{(*)}$	1	1	1
17	GA	0.8565	0.8344	1
	KLA	0.5820	0.7113	0.9896
	CEA$^{(*)}$	1	1	1
No. ingredients	5			
No. points	Algorithm	RD	RUD	VD
15	GA	0.8909	0.8217	1
	KLA	0.1590	0.3534	0.5196
	CEA$^{(*)}$	1	1	1
30	GA	0.7710	0.7857	1
	KLA	0.4245	0.7371	0.9252
	CEA$^{(*)}$	1	1	1

Supplementary Material D

Table 3: *Relative D- and I-efficiencies for model (8) under different scenarios (RD, RUD and VD).* $^{(*)}$ *indicates that the obtained result does not depend on a set of candidate points. E denotes the design obtained by the practitioners.*

No. ingredients	3	*D*-optimality			*I*-optimality		
No. points	Algorithm	RD	RUD	VD	RD	RUD	VD
7	GA	0.9023	0.9511	1	0.9887	0.9929	1
	KLA	0.7004	0.8294	1	0.8028	0.9480	0.9916
	E$^{(*)}$	0.8669	0.8669	0.8669			
12	GA	0.9557	0.9538	1	0.9998	0.9559	1
	KLA	0.7488	0.8744	0.9991	0.8194	0.9184	0.9993
	MA$^{(*)}$	0.9979	0.9979	0.9979	0.9100	0.9100	0.9100
18	GA	0.9869	0.9610	1	0.9785	0.9687	1
	KLA	0.7610	0.9043	0.9931	0.9081	0.9582	0.9986
	MA$^{(*)}$	0.9606	0.9606	0.9606	0.9711	0.9711	0.9711
No. ingredients	5	*D*-optimality			*I*-optimality		
No. points	Algorithm	RD	RUD	VD	RD	RUD	VD
15	GA	0.6365	0.9198	1	0.6774	0.8576	1
	KLA	0.2106	0.4728	0.9655	0.1243	0.3520	0.5082
30	GA	0.6452	0.8764	1	0.6353	0.7511	1
	KLA	0.2751	0.4863	0.9613	0.2546	0.5658	0.8550

Table 4: *I-optimal designs obtained with GA and KLA for model (8), five-ingredient mixtures and $N = 15$ runs.*

GA					KLA				
p1	p2	p3	p4	p5	p1	p2	p3	p4	p5
0.0076	0.9254	0.0240	0.0008	0.0422	0.5000	0.4667	0.0000	0.0000	0.0333
0.0046	0.0317	0.0001	0.0304	0.9331	0.0000	0.0000	0.3000	0.3667	0.3333
0.0332	0.0032	0.0302	0.9332	0.0001	0.0000	0.0000	0.0333	0.9333	0.0333
0.0001	0.3332	0.3332	0.0001	0.3333	0.0000	0.3333	0.3333	0.0000	0.3333
0.0713	0.0033	0.8447	0.0101	0.0706	0.0000	0.9667	0.0000	0.0333	0.0000
0.3327	0.0015	0.3195	0.3131	0.0333	0.9667	0.0000	0.0000	0.0000	0.0333
0.1174	0.4035	0.3864	0.0919	0.0007	0.4667	0.0000	0.5333	0.0000	0.0000
0.4693	0.0045	0.0325	0.0010	0.4927	0.0000	0.0000	0.7000	0.2333	0.0667
0.1889	0.0414	0.0224	0.4237	0.3236	0.5000	0.1333	0.0000	0.2333	0.1333
0.0003	0.0347	0.0384	0.4653	0.4613	0.2000	0.2000	0.0000	0.2000	0.4000
0.1557	0.2985	0.0548	0.0339	0.4571	0.2333	0.4667	0.2667	0.0333	0.0000
0.4288	0.2673	0.0125	0.2765	0.0150	0.5667	0.0667	0.1000	0.0667	0.2000
0.0011	0.4905	0.0020	0.4591	0.0473	0.0333	0.0333	0.1667	0.0667	0.7000
0.1054	0.0246	0.2378	0.0457	0.5865	0.1667	0.0000	0.2333	0.5667	0.0333
0.0022	0.0379	0.4696	0.4552	0.0351	0.0333	0.4000	0.1333	0.4333	0.0000

Supplementary Material E

Table 5: *Relative D-efficiencies for model (9) under different scenarios (RRD, SEVD and EVD).* $^{(*)}$ *indicates that the obtained result does not depend on a set of candidate points.* $^{(\star)}$ *means that the* 4*-th and* 5*-th ingredient were added to the problem without constraints, whereas* $^{(\star\star)}$ *denotes that these were constrained.*

No. ingredients	3			
No. points	Algorithm	RRD	SEVD	EVD
6	GA	1	1	1
	KLA	0.8937	0.6406	1
	CEA$^{(*)}$	1	1	1
12	GA	1	1	1
	KLA	0.8112	0.6582	1
	CEA$^{(*)}$	1	1	1
No. ingredients	5			
No. points	Algorithm	RRD	SEVD	EVD
15$^{(\star)}$	GA	1	0.9828	1
	KLA	0.5150	0.6008	0.6953
	CEA$^{(*)}$	1	1	1
15$^{(\star\star)}$	GA	1	1	1
	KLA	0.5000	0.6250	0.750
	CEA$^{(*)}$	1	1	1

Table 6: *Relative I-efficiencies for I-optimality and model (9) under different scenarios (RRD, SEVD and EVD).* $^{(*)}$ *indicates that the obtained result does not depend on a set of candidate points.* $^{(\star)}$ *means that the* 4*-th and* 5*-th ingredient were added to the problem without constraints, whereas* $^{(\star\star)}$ *denotes that these were constrained.*

No. ingredients	3			
No. points	Algorithm	RRD	SEVD	EVD
12	GA	1	0.9933	0.9935
	CEA$^{(*)}$	0.6474	0.6474	0.6474
No. ingredients	5			
No. points	Algorithm	RRD	SEVD	EVD
30$^{(\star)}$	GA	0.9404	1	0.9407
	CEA$^{(*)}$	0.4991	0.4991	0.4991
30$^{(\star\star)}$	GA	0.8790	0.8931	1
	CEA$^{(*)}$	0.5207	0.5207	0.5207

Supplementary Material F

Table 7: *Relative D-efficiencies for model (10) under different scenarios (RRD, SEVD and EVD).* $^{(*)}$ *indicates that the obtained result does not depend on a set of candidate points.*

No. ingredients	4			
No. points	Algorithm	RRD	SEVD	EVD
14	GA	0.9440	1	0.9120
	KLA	0.2590	0.3090	0.4390
	CEA$^{(*)}$	0.5594	0.5594	0.5594
20	GA	1	0.95	0.85
	KLA	0.3100	0.3900	0.48
	CEA$^{(*)}$	0.5348	0.5348	0.5348

Table 8: *Relative I-efficiencies for I-optimality and model (10) under different scenarios (RRD, SEVD and EVD).* $^{(*)}$ *indicates that the obtained result does not depend on a set of candidate points. E denotes the design obtained by the practitioners.*

No. ingredients	4			
No. points	Algorithm	RRD	SEVD	EVD
20	GA	0.8190	1	0.8863
	CEA$^{(*)}$	0.5823	0.5823	0.5823
	E$^{(*)}$	0.0451	0.0451	0.0451