EXPERIMENTS WITH MIXTURES

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1. Introduction

The subject of design of experiments deals with the statistical methodology required for making the inferences about the treatment effects on the basis of responses (univariate or multivariate) collected through the planned experiments. To deal with the evaluation and analysis of a system of variables, the experiments involving several factors simultaneously are being conducted in agricultural, horticulture and allied sciences. Data from experiments with levels of combinations of one or more factors as treatments are normally investigated to compare the level effects of the factors and also their interactions. Though such investigations are useful to have objective assessment of the effects of the levels actually tried in the experiment, this seems to be inadequate, as these do not throw much light on the possible effect(s) of intervening levels or their combinations. In these situations, if all the factors are quantitative in nature, it is natural to think the response as a function of the factors of levels and data from quantitative factorial experiments can be used to fit the response surface over the region of interest. Response surface designs are developed to handle such situations. In response surface designs the levels of factors are independent and the quantity of the inputs (levels) are fixed by the experimenter to obtain the region of optimum response. In another type of experiments called *experiments with mixtures*, the response depends upon the proportions of the input ingredients used *i e*. in mixture experiments we study the performance of various mixtures formed by mixing two or more ingredients. The relative amount of a particular component with respect to the total amount of the mixture is known as the proportion or fraction of that component in the mixture. Hence, the proportions of mixture components in a combination add up to unity. By virtue of this, the totality of the unrestricted factor space of p-dimension reduces to a p-1 dimensional.

The designs for mixture experiments have been extensively used in agricultural/ industrial experiments. Some of the situations in which these designs could be advantageously used are: split application of fertilizers, intercropping experiments where the interest of experimenter is to find best crop mixtures, sensory evaluation experiments for making the agricultural and animal products, preparation of fertilizers, insecticides/ pesticides mixtures for optimum response, feeding trials in animal nutritional experiments. Mixture experiments can be used in with unrestricted region, with restricted region having upper and/or lower bounds, with process variable(s). Some of the experimental situations where mixture experiments are quite useful in agricultural sciences are:

Experimental Situation 1: Consider two chemical pesticides (P_1 and P_2) each of which could be used for killing mites on strawberry plants by spraying the pesticides on plants. One wished to determine if the combination of P_1 and P_2 exist that are more effective than either of pesticide by itself. To remove the effect of the amount of pesticide applied, the experiment consists of rank-fixing five different combinations of P_1 and P_2 where all blends have the

same volume. Expressed in percentage of this fixed volume, the five mixes are (P_1 : P_2) = (0.90 : 0.10), (0.75 : 0.25), (0.50 : 0.50), (0.25 : 0.75), (0.10 : 0.90). Then, if the same amount of each of the five different blends is sprayed on five different groups of plants, where each group of plants possesses the same number of mites and after a specific period of time as percentage of mites killed is noted from one group to next. Thus, the difference in effectiveness of five blends of P_1 and P_2 is a mixture of relative proportions of P_1 and P_2 in the blends and is not a function of the amount of pesticide applied.

Experimental Situation 2: In fertilizer trials, the experimenter may be interested in obtaining the optimum time of applications of fixed dose of fertilizer to a crop. Treatments here consist of applying fixed quantity of fertilizer in splits at different crop growth stages. For example, an experiment was conducted to study the effect of nitrogen applied in splits at different crop growth stages on yield of paddy crop at Rice Research Station, Behrampur (Orissa) in 1971. The treatments consisted of applying 100 kg. of nitrogen/hectare as Calcium Ammonium Nitrate at different crop growth stages viz. planting, tillering, panicle initiation and booting. Different combinations of treatments tried are given in Table 1.

Table 1: Split application of fertilizer to crop at different crop growth stages

Treatment	Crop growth Stage			
	Planting	**Tillering**	**Penicle initiation**	**Booting**
T_0	0	0	0	0
T_1	1.00	0	0	0
T_2	0.75	0.25	0	0
T_3	0.75	0	0.25	0
T_4	0.75	0	0	0.25
T_5	0.50	0.25	0.25	0
T_6	0.50	0	0.25	0.25
T_7	0.50	0.25	0	0.25
T_8	0.25	0.50	0.25	0
T_9	0.25	0.25	0.25	0.25
T_{10}	0	0.50	0.50	0
T_{11}	0	0.50	0.25	0.25

Source: Summarization of experimental data on paddy crop for Eastern Region 1966-75, IASRI publication, pp.158-159.

Experimental Situation 3: An experiment was conducted to study the effect of application of Urea and Ammonium Sulphate in varying proportions on yield and quality of bidi tobacco at Bidi Tobacco Research Station, Anand (Gujrat). The five treatments tried were application of fixed quantity (180 kg/ha) of nitrogen in proportions are given in Table-2.

Table 2: Split application of nitrogen obtained from two different sources

Treatment No.	Urea		Ammonium Sulphate	
	Quantity (N kg/ha)	Proportion	Quantity (N kg/ha)	Proportion
T1	180	1.0	0	0
T2	135	0.75	45	0.25
T3	90	0.50	90	0.50
T4	45	0.25	135	0.75
T5	0	0	180	0

This experiment was conducted using randomized complete block design with six replications. The main aim of the experiment was to obtain the optimum split of nitrogen (amount of N from urea and Ammonium Sulphate) that gives the maximum yield.

Experimental Situation 4: A sensory experiment was carried out in Post-Harvest Technology unit of Division of Fruits and Horticultural, IARI, New Delhi, to study feasibility of blending of fruit juicepulp. Evaluation of juicepulp and their processing was done as per the standard procedure. Juicepulp of mango (Dushehari), pineapple (kew), aonla (Desi), lime (Kagzi) and grapes (Pusa navarang) was blended with two juicepulp (lime-aonla, mango-pineapple, grape-mango and grape-pineapple) in different ratios [(0 : 1.00), (0.05 : 0.95), (0.10 : 0.90), (0.15 : 0.85), (0.20 : 0.80), (0.25 : 0.75), (0.50 : 0.50), (1.00 : 0), (0.95 : 0.05), (0.90 : 0.10), (0.85 : 0.15), (0.80 : 0.20), (0.75 : 0.25)]. The different blended juicepulp were used for preparation ready to serve (RTS) beverages. The RTS beverages were evaluated adopting nine point Hedonic scale. It was found that the scores for overall sensory quality were higher for mango-pineapple and grape-pineapple blends and lower for lime-blending ratio of fruit pulp for the beverages on a commercial scale.

Experimental Situation 5: An experiment was conducted at Dryland Research Project, Dr. Panjabrao Deshmukh Krishi Vidyapeeth, Akola, to find out the optimum row spacing in sorghum + green gram intercropping. There were five row ratios of sorghum and green gram, viz. 1:0, 1:2, 1:1, 2:1 and 0:1and two row spacing, viz. 30 cm and 45 cm, forming 10 treatment combinations. Treatment details are given in Table – 2.

Table 3: Proportion of two crops with two row spacing

Treatment	Crops	Row Ratio	Row spacing (cm)
T_1	Sorghum + Green gram	1:2	30
T_2	Sorghum + Green gram	1:1	30
T_3	Sorghum + Green gram	2:1	30
T_4	Sorghum + Green gram	1:2	45
T_5	Sorghum + Green gram	1:1	45
T_6	Sorghum + Green gram	2:1	45
T_7	Sole Sorghum	1:0	30
T_8	Sole Sorghum	1:0	45
T_9	Sole Green gram	0:1	30
T_{10}	Sole Green gram	0:1	45

In above experiment with crop mixtures, we want to study the response (which may be the total yield in money value) with two-row spacing. We are thus extending the crop mixture experiment by trying it at different levels of row spacing. The new factor row spacing (in present case), which does not form any component of mixture, is called the *process variables* of the experiment and the crops, as usual, are called the *mixture variables*.

In mixture problems the main considerations connected with the exploration of response surface over simplex region are:

(i) The choice of the proper model to approximate the surface over the region of interest
(ii) The testing of adequacy of the model in representing the response surface and
(iii) A suitable design for collecting the observations, fitting the model and testing the adequacy of fit.

An excellent account of work done on experiments with mixtures is given in the book by and Khuri and Cornell (1987) and Cornell (2002). Also the applications of mixture designs with their analysis are well documented in Murthy and Das (1968), Batra *et al.*(1999), Deka *et al.*(2001) and Dhekle *et al.*(2003).

2. Mixture Experiments Models

A mixture experiment involves the study of performance of various mixture formed by mixing two or more components called ingredients. Let x_i represents the proportion of i^{th} ingredient in a mixture. Then evidently

$$0 \le x_i \le 1 \qquad i = 1\ (1)\ q \tag{2.1}$$

and

$$\sum_{i=1}^{q} x_i = 1 \tag{2.2}$$

where q is the number of components. In mixture experiments, the factors are ingredients of a mixture and their levels are not independent.

Thus mixture experiments are such, wherein the property studied, *viz* the response (dependent variable) depends upon the proportions of components (independent variables) present but not on the amount of the mixture. The relative amount of a particular component with respect to the total amount of the mixture is known as the proportion or fraction of that component in the mixture. Hence, the proportions of mixture components are formed by blending together non-negative quantities of the various ingredients and the sum of non-negative component proportions is fixed for all design points and taken as unity. If an individual proportion x_i is unity, then mixture is called pure blend, if two of the $x_i>0$, then blend is called binary and so on. Let us assume that

$$\eta=\phi(x_1,...,x_q) + e \tag{2.3}$$

be a functional relationship between the response value (η) which is dependent on the proportions $x_1,...,x_q$ of the components of the mixture . The problem of associating the response variable (y) with ingredient compositions centers on determining the mathematical equation that describes the function ϕ (.). In general, the polynomial functions are used to represent $\phi(x_{1,...,}x_q)$, however , the polynomial functions are different from the polynomial

equations considered for the usual regression problems. Applying the condition (2.2) in standard polynomials, Scheffe (1958, 1963) derived the polynomials for mixture problems and termed as canonical polynomials.

For understanding the procedure of obtaining canonical polynomials from standard polynomials we shall, consider a two component mixture problem, where mixture proportions for the two components denoted by x_1 and x_2 such that $x_1 + x_2 = 1$. For the polynomial of degree one, in standard notations,

$$\eta = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + e \qquad (2.4)$$

where β_0, β_1 and β_2 are unknown coefficients. Now applying condition (2.2), we have

$$\eta = \beta_0(x_1 + x_2) + \beta_1 x_1 + \beta_2 x_2 + e$$

$$\eta = (\beta_0 + \beta_1)x_1 + (\beta_0 + \beta_2)x_2 + e$$

$$\eta = \beta_1' x_1 + \beta_2' x_2 + e \qquad (2.5)$$

similarly, for the seconds degree polynomial

$$\eta = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_{12} x_1 x_2 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + e \qquad (2.6)$$

replacing x_1^2 by $x_1(1-x_2)$; x_2^2 by $x_2(1-x_1)$, we get

$$\eta = \beta_1' x_1' + \beta_2' x_2' + \beta_{12}' x_1 x_2 + e \qquad (2.7)$$

where

$$\beta_1' = \beta_0 + \beta_1 + \beta_{11}\,;\ \beta_2' = \beta_0 + \beta_2 + \beta_{22}\,;\ \beta_{12}' = \beta_{12} - \beta_{11} - \beta_{22}$$

From (2.4), intercept term and from (2.6), intercept term, pure quadratic terms $\beta_{11} x_1^2$ and $\beta_{22} x_2^2$ have been eliminated. Thus the canonical polynomials have fewer terms as compared to ordinary standard polynomials. Further, higher degree canonical polynomials can similarly be obtained. In general linear and quadratic polynomials for q component system are:

Linear : $$\eta = \sum_{i=1}^{q} \beta_i x_i + e \qquad (2.8)$$

Quadratic : $$\eta = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q}\sum^{q} \beta_{ij} x_i x_j + e \qquad (2.9)$$

The quadratic model (2.9) is full quadratic. If we do not take some of the cross product terms in the model, then, we have termed it as restricted quadratic model.

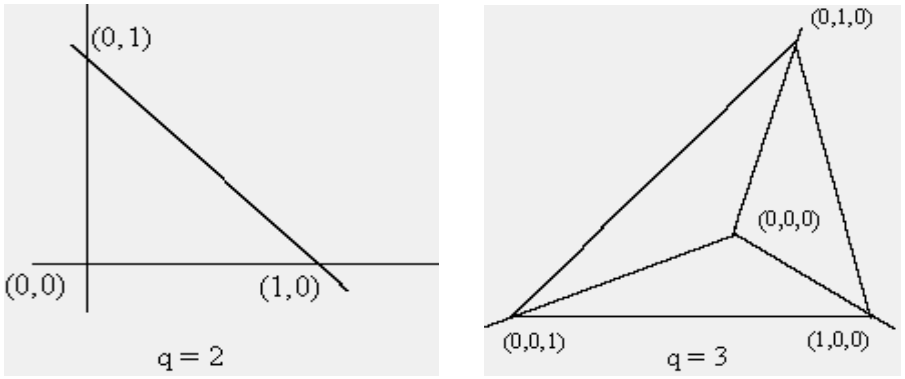
The terms in canonical polynomials (linear, quadratic and restricted quadratic) have simple interpretations. β_i (i=1(1)q) represents the expected response to the pure mixture x_i=1, x_j=0 $\forall$ j, i=1(1)q and defines the height of the mixture surface at the simplex vertex denoted by x_i=1. The portion $\sum_{i=1}^{q} \beta_i x_i$ of each of the models is called linear blending portion and if the blending of components is strictly additive then linear canonical polynomials are most appropriate representation of the surface.

When there is a curvature in the mixture surface owing to non-linear (often called synergistic and antagonistic) blending between pairs of components, then, canonical polynomial of degree two or more are fitted as representation of the surface. The parameters β_{ij} in (2.9)

represents the deviation from surface represented by (2.7). If sign of β_{ij} is positive, the components i and j are synergistic and if negative they have antagonistic behavior.

3. Design Points for Mixture Experiments

In mixture experiments, the experimental region or factor space of interest is defined by value of x_i's (mixture proportion) in a regular *q*-1 dimensional simplex. Here, *q* denotes the number of ingredients of the mixture. For $q = 2$ components, the factor space is a straight line and for $q=3$, it is an equilateral triangle. For four components, it is a regular tetrahedron. Fig. 3.1 denotes the factor space for $q = 2$ and $q = 3$ mixture experiments. The coordinates viz. (0,1), (1,0) for $q = 2$ and (1,0,0), (0,1,0), (0,0,1), (1/2,1/2,0), (1/3,1/3,1/3) for $q = 3$, etc. are called mixture points and the coordinate system for mixture proportions is called simplex coordinate system. Scheffe (1958,1963) gave simplex lattice and simplex centroid designs for experiments with mixtures. We describe these designs briefly, for other useful designs for mixture experiments one may refer to Cornell(2002).



(Fig. 3.1)

3.1 Simplex Lattice Designs

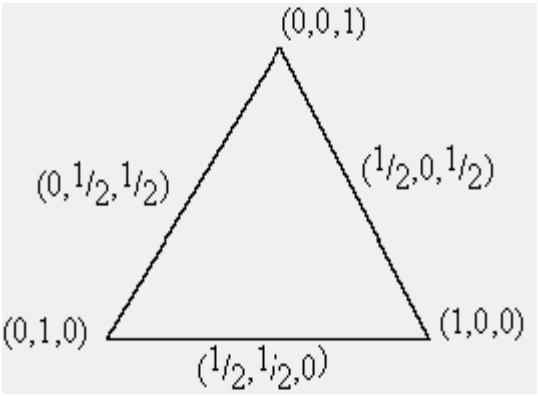
For a q-component system, a (*q*,*m*) simplex lattice consists of points defined by following coordinate settings: the proportions assumed by each component takes the m+1 equally spaced values from 0 to 1 viz.

$$x_i = 0, 1/m, 2/m, \ldots, 1; \quad \text{for } i = 1, \ldots, q \qquad (3.1)$$

All possible combinations of x_i`s (3.1) are considered as design points. For example, when q=3 and m=2, each component can take the proportions x_i=0,1/2,1; $\forall$ i=1,2,3. Then, (3.2) simplex lattice consists of six points on the boundary of triangular factor space,

x_1	x_2	x_3
1	0	0
0	1	0
0	0	1
1/2	1/2	0
1/2	0	1/2
0	1/2	1/2

The three vertices (1,0,0), (0,1,0), (0,0,1) represent the individual components (pure blends) while the points (1/2,1/2,0), (1/2,0,1/2), (0,1/2,1/2) represent the binary blends or two-component mixtures and are located at the mid-points of the three sides of an equilateral triangle (fig.3.2)



(Fig. 3.2)

The number of component combinations that belong to the (q,m) simplex lattice is ${}^{q+m-1}C_m$

3.2 Simplex Centroid Design

These designs were introduced by Scheffe(1963). In a q-component simplex centroid design, the number of points is 2^q -1. The design points correspond to the *q*-permutations of (1,0,0,0,.....,0), ${}^qC_2$ permutations of (1/2,1/2,0,0,....,0), the ${}^qC_3$ permutations of (1/3,1/3,1/3,0,...,0),..., and the centroid point(1/q,1/q,...,1/q). a four-component simplex centroid design consists of 2^4-1 = 15 design points and is given in the following Table.

x_1	x_2	x_3	x_4
1	0	0	0
0	1	0	0
0	0	1	0
0	0	0	1
1/2	1/2	0	0
1/2	0	0	1/2
1/2	0	1/2	0
0	1/2	0	1/2
0	1/2	1/2	0
1/3	1/3	0	1/3
1/3	1/3	1/3	0
1/3	0	1/3	1/3
0	1/3	1/3	1/3
1/4	1/4	1/4	1/4

4. Analysis of Mixture Experiments

4.1 Least Squares and ANOVA

For a general linear model

$$\mathbf{y} = \mathbf{X}\boldsymbol{\beta} + \mathbf{e} \tag{4.1}$$

where $\mathbf{y}$ is an n x1 vector of observations is n×p matrix whose elements are the mixture component proportions and functions of the component proportions, $\boldsymbol{\beta}$ is a p×1 vector of unknown parameters and $\boldsymbol{\varepsilon}$ is a vector of random errors distributed as $N(0,\sigma^2)$ i.e. $E(\boldsymbol{\varepsilon}) = \mathbf{0}$ and $D(\boldsymbol{\varepsilon}) = \sigma^2\mathbf{I}_n$. Corresponding to model (2.3), we have $p = q$ in (2.8) and for (2.9), $p = q(q+1)$. The normal equations are

$$\mathbf{X}'\mathbf{X}\,\boldsymbol{\beta} = \mathbf{X}'\mathbf{y} \tag{4.2}$$

and least squares estimates of $\boldsymbol{\beta}$`s are

$$\hat{\boldsymbol{\beta}} = (\mathbf{X}'\mathbf{X})^{-1}\mathbf{X}'\mathbf{y} \tag{4.3}$$

ANOVA

Source of variation	Degrees of freedom	Sum of square	Mean square	F-value
Regression (fitted model)	p - 1	$SSR = \hat{\boldsymbol{\beta}}\mathbf{X}'\mathbf{y} - (\mathbf{1}'\mathbf{y})^2/n = \sum_{u=1}^{n}(\hat{y}_u - \bar{y})^2$	MSR = SSR/(p-1)	MSR/ MSE
Error	n - p	$SSE = \hat{\boldsymbol{\beta}}\mathbf{X}'\mathbf{y} - (\mathbf{1}'\mathbf{y})^2/n = \sum_{u=1}^{N}(y_u - \hat{y}_u)^2$	MSE = SSE/(n-p)	
Total	$n-1$	$SST = \mathbf{y}'\mathbf{y} - (\mathbf{1}'\mathbf{y})^2/n = \sum_{u=1}^{n}(y_u - \hat{y})^2$		

It is important to note here that from canonical polynomials, it seems that for ANOVA, the regression sum of square (SSR) and total sum of square (SST) should be uncorrected. But for canonical polynomials, these should be corrected ones. For more details, one may refer to Marquardt and Snee(1974).

It has been widely advocated that regression model should be fitted on average data [see e.g. Gomez and Gomez (1976)], however, to make use of replicated data available, fit the model on replicated data ignoring the blocking whenever block differences are non-significant and adjusted yield to block means whenever block differences are significant.

4.2 Model Selection Criterion

The fitted models are tested for lack of fit using the F statistics

$$F = \frac{SS_{lof}/(v-p)}{SS_{pe}/(N-p)}$$

where n is the total number of observations, v is the number of distinct treatments and p is the number of terms included in the model. SS_{pe} (sum of squares due to pure errors) is calculated in the following manner. Denote the l^{th} observation at the u^{th} design point by y_{lu}, where

l=1,....., r_u>=1, u=1,.....v . Define $\mathbf{y_u}$ to be average of the r_u observation at the uth design point. Then, the sum of squares for pure error is

$$SS_{pe} = \sum_{u=1}^{v} \sum_{l=1}^{r_u} (y_{lu} - \bar{y}_u)^2$$

the sum of squares due to lack of fit is then obtained by subtraction

$$SS_{lof} = SSE - SS_{pe} \qquad (4.4)$$

The model then is selected on the basis of non-significance of lack of fit. However, if lack of fit was non-significant in more than one case, the model is selected on the basis of smallest value of standard error of estimates of y_3 i.e. smallest mean square error. In literature, however other statistic R^2 (multiple correlation coefficient), R_a^2 (adjusted correlation coefficient) and PRESS predicted residual sum of squares) have been considered for model selection. Montgomery and Peck (1982) have recommended the use of standard error of estimate for model selection criterion if the possible use of regression is data description. Therefore, the model is selected on the basis of standard error of estimate and other statistic viz. R^2, R_a^2 and PRESS have also been given for the model.

As averaging of data helps in smothering the data and reduces the error variance and hence, final model selected is on the basis of average data. It is important to note here that the estimates of coefficient of various terms in the model are same for replicated data as well as for average data. The standard error of estimates of coefficients for average data models is smaller. It is however, interesting to note here that model selected from replicated data analysis and average data are same including the terms in the model.

Illustration 1: An experiment was conducted at Agril. Station, Ragalu (A.P.) to determine the optimum time of application of 100 Kg/ha of N to Paddy crop. The fertilizer was applied at three crop growth stage viz Basal (x1),Tillering (x2), Panicle initiation (x3). Experiments was conducted through RCB design in 4 replications with fallowing set of treatments. The average yield of Paddy obtained is as given below:

x_1	x_2	x_3	**Av. yield (q/ha)**
1	0	0	11.20
3/4	1/4	0	12.33
3/4	0	1/4	14.04
1/2	1/2	0	16.41
1/2	0	1/2	18.26
1/2	1/4	1/4	16.89
1/4	3/4	0	18.22
1/4	1/2	1/4	19.42
0	1	0	20.61
0	3/4	1/4	19.93
0	1/2	1/2	20.77

Fit a suitable second order polynomial to the above data.

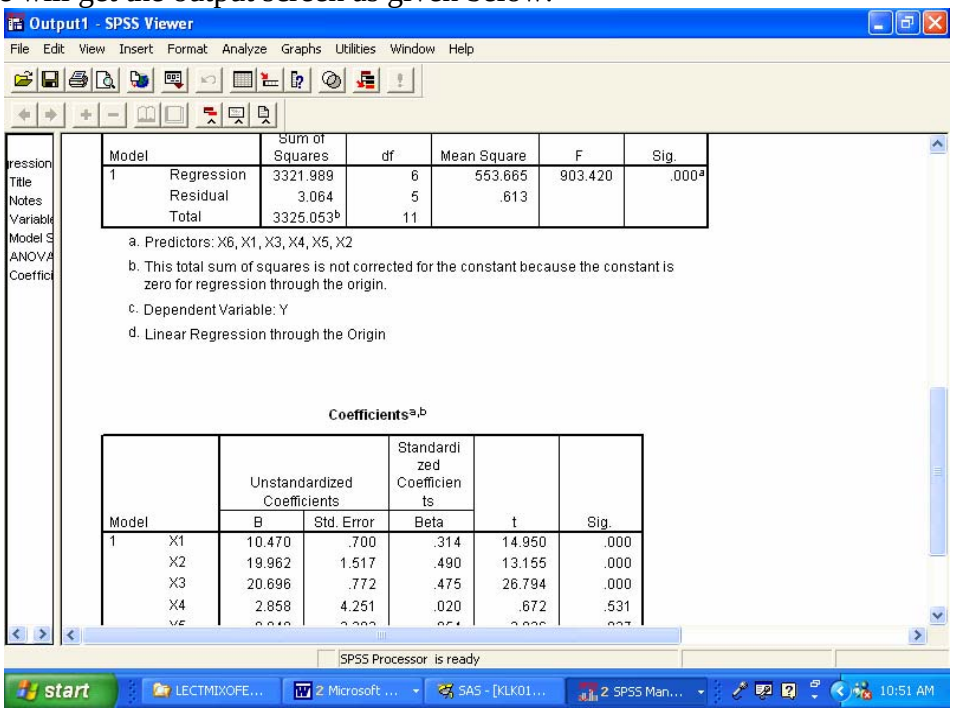
The data can be analysed by using any authentic software. Here we show by using the software SPSS and SAS.

A) By using SPSS

- First we enter the data in spreadsheet of SPSS by using the variables x_1, x_2, x_3, and yld
- Now if we have To use second order model, we first compute quadratic terms x_1x_2, x_1x_3 and $x_2\ x_3$ by using following commands
- Transform → Compute → Box → button [Target variable(variable where the product term is to store)] → button [Numeric expression(write the formula)] →Box(Change existing →OK.
- Now for analysis, we follow as

- Analyze → Regression → Linear → button [Put y under Dependent variable] → button [Put x_1, x_2, x_3, x_{12}, x_{13}, x_{23} under independent] → Options → button [Tick off (Box) Include constant in equation] → Continue → OK

- We will get the output screen as given below:



B) By using SAS

Now we give the steps used while analyzing the data using SAS software.
First we analyze the data for four years separately using **proc reg.**

```
data mixexp;
input x1, x2, x3 y;
x4=x1*x2;
x5=x1*x3;
x6=x2*x3;
 cards;
DATA;
proc reg;
model yld=x1 x2 x3/noint ss1 ss2 p;
proc reg;
model yld=x1 x2 x3 x4 x5 x6/noint ss1 ss2 p;
 run;
```

5. Designs for Restricted Region

In many mixture experiments, it is not possible to explore the whole range $0 \le x_i \le 1$ with all components. This is because one may require that at least a certain proportion of ($0 \le L_i \le x_i$) of component i be present in all blends, thus eliminating the case where $x_i = 0$ or one may insist that component j contributes at most a proportion ($x_j \le U_j \le 1$). An experimenter may thus put either a lower bound or upper bound or both to one or more than one component. If only lower bounds are placed on the proportion of one or more components which are of the form $x_i \ge L_i \ge 0$; i=1,...,q and $\sum_{i=1}^{q} L_i = L<1$. We define the Pseudo Components.

$$x_i = \frac{x_i - Li}{1-L}; \quad \sum_{i=1}^{q} x_i' = 1.$$

We set up a design in x_i' which are than mapped back to provide setting in the original components. The data are then collected on the blends in original components and are used to fit a model. In case both upper (U_i) and lower (L_i) bounds are placed on component proportion viz.$0 \le L_i \le x_i \le U_i \le 1$. The feasible mixture of blends is a convex polyhedron. The vertices of a polyhedron are candidates of design points and can be obtained through XVERT1 algorithm of Nigam *et al.* (1983).

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