GAB 模式應用於農產品等溫水份物性之評估

Evaluation of the GAB Equation for the Isotherms of Agricultural Products

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摘 要

在此研究中以具有三參數之 GAB 模式評估其於數種農產品等溫水份數據之適用性。此模式僅能用於表達固定溫度下含水率與平衡相對濕度（或稱水活性）之關係，以水活性爲非獨立變數時，此模式之型式十分複雜。由非線性迴歸之殘差圖顯示此模式並不適用含澱粉質之農產品。對大多數的農產品而言，此模式之三參數與溫度並未具有 Arrhenius 型關係，在參數項加入溫度函數則使模式增加更多參數數目而減少適用性，因此此模式並不適用於農產品之等溫水份物性。

關鍵詞：GAB 模式、等溫水份線

ABSTRACT

The three parameter GAB equation was used to evaluate the fitting-agreement of the sorption data for several agricultural products. This model only indicates the relation between moisture content and equilibrium relative humidity (or Water activity, Aw) at fixed temperature. As the ERH values serve as the dependent variable, the equation become a very complex form. The residual plots of this model show that it cannot fit well for starchy products. The parameters of GAB equation for the most products did not have the Arrhenius-type relationship with temperature. To incorporate the temperature term into the parameters gives the model become more parameters and less usefulness. It cannot be recognized as an adequate sorption model for agricultural products.

Key words: GAB equation, Isotherms

INTRODUCTION

Equilibrium moisture content (EMC)-equilibrium relative humidity (ERH) relationship are essential factors to design drying, handling, storing, mixing, packaging and modeling seed longevity for agricultural products. The ERH value was called water activity (Aw) for food sciences and engineer. An adequate equation mathematically describes the relationship between the ERH (or Aw), moisture content and temperature is very useful for simulation work of processing. Many theoretical, semi-theoretical, and empirical isotherm equations have been developed to model the relationship.

Chen and Morey (1989) evaluated the fitting-ability of four ERH/EMC equation for grains and seeds. The data include thirty-five data sets comprising 18 crops. The Modified-Henderson and Chung-Pfost equations are good models for starchy grains and fibrous materials. The Modified-Halsey equation is good model for high oil and protein products. The Modified-Oswin equation fits the data for selected crops.

An equation, derived independently by Guggenhein, Anderson, and de Boer from a physical adsorption model related to the BET theory, has been adopted widely among food scientists recently (Van den Berg, 1984, 1985).

The GAB equation has the following form:

$$M = \frac{Mo*K*C*Aw}{(1-K*Aw)(1-K*Aw+c*K*Aw)} \qquad (1)$$

Where (2)

$C = Cl*Exp(Hm-Hn)/RT)$ (2)
$K = Kl*Exp(Hl-Hn)/RT)$ (3)
Aw = Water activity
Mo = Molecular value
Hm = Molar sorption enthalpy of the monolayer
Hn = Molar sorption enthalpy of the multilayers on the top of the monolayer.
Hl = Molar sorption enthalpy of the bulk liquid , C1 and K1 are constants.

The GAB model describes the moisture content as a function of temperature and water activity. If water activity is chosen as a funtion of moisture content and temperature, this equation transforms to a form given by,

$$Aw = \frac{2 + (Mo/M-1)C - [(2+(Mo/M-1)C)^{2.0} - 4(1-C)]^{0.5}}{2*K*(1-C)} \qquad (4)$$

The major advantages of GAB model, according to Van den Berg (1984), are:

1. It has a theoretical background that is refined from BET theory.
2. It describes sorption behavior of nearly all foods from zero to 0.9 Aw.
3. It has a relatively simply mathematical form with only three parameters.
4. Its parameters have a physical meaning in terms of the sorption process.
5. It is able to describe some temperature effects of isotherms by Arrhenisu-type equations.

After transformation, the GAB equation has an equivalent form to the Hailwood and Horrobin equation (1946).

$$\frac{Aw}{M} = a1 + a2*Aw + a3*Aw^2 \qquad (5)$$

The equivalent of parameters for GAB equation are:

$$K = \frac{\sqrt{a_2^2 - 4*a_1*a_3} - a_2}{2a_1} \qquad (6)$$

$$C = \frac{a_2}{a_1*K} + 2 \qquad (7)$$

$$Mo = \frac{1}{a_1*K*C} \qquad (8)$$

With this simple algebraic from, the parameters of the nonlinear GAB equation can be calculated easily by linear multiple regression analysis. This method was used by Bizot (1984) to construct sorption isotherms and by Wesier (1985) to find the parameters for four food materials. Schar (1985) compared the method of obtaining the parameters and found that the values depended on the type of regression procedure used.

Lomauro et al. (1985) used the GAB equation and three two-parameter equations to evaluate the goodness of fit for 163 sorption isotherms. An equation having a mean relative deviation models of less than 5 was considered to represent the good fitting-ability. They found the GAB equation could present the best accuracy.

After using the GAB model to construct sorption isotherms, Bizot (1984) proposed that the equation with physically meaningful coefficients could fit data very well up to 0.9 Aw in many cases. However, he also emphasized the GAB model had some limitations. The model did not apply to all shapes of isotherms. As the temperature changed, the fit might be less satisfactory. The model did not fit the more crystalline forms of starch at Aw range 0.6-0.7.

Because the GAB equation does not include the temperature term. It only can present the relationship between Aw and moisture content at fixed temperature Iglesias and Chirife (1984) represented the monolayer value as an Arrhenius relation with temperature.

$$M = M1*Exp(-M2/T) \tag{9}$$

Using this equation to express the temperature dependence of Mo and adopted the equation (2) and (3) to express the temperature relationship of K and C, Weiseser (1985) showed that the GAB equation with temperature coefficients is suitable for describing the influence of temperature on the sorption behavior of four food components in the range of 25-80 C. The same method was applied to ground roasted coffee for five temperatures (Weisser, 1986).

Conversely, Cencturk et al. (1986) studied the sorption isotherms of wild rice and found that the three parameters wre not a function of temperature.

Recently, the GAB equation began to be mentioned by agriculture engineers. Bakker-Arkema (1985) recommended this equation for modeling the simulation working of dryer. Mazza et al. (1990, 1991) estimated the coefficients of this equation for the isotherm of flax seed and Sunflower seeds, kernels, and hulls.

The objective of this study is to evaluate the fitting ability of the GAB equation for the isotherm data of agricultural products.

MATERIALS AND METHODS

1. Data collection

The sorption isotherms of several temperatures for eight cereal grains and oil seeds were selected to find the parameters of the GAB equation (Table 1). The known parameter for five foodstuffs (Table 2) are used to study the Arrhenius relationship between the GAB parameter and temperature.

2. Data analysis

GAB equation is a nonlinear, three-parameter equation. A program, "GAB", written by GW-BASIC language was used to estimate the parameters and statistics.

The criteria for comparing were standard error of the estimated value (S.E.), mean relative percent error (P) and residual plots. The clean pattern appeared to the residual plots indicates that the model fails to explain the variation in the data.

The standard error of the estimate values was defined as:

$$S.E. = \sqrt{\frac{\Sigma(Y - Y')^2}{df}} \tag{10}$$

where Y = The measured values
Y' = The predicted values
df = Degrees of freedom of the model

The mean relative percent error was:

$$P = \frac{100}{N} * \Sigma \frac{|Y - Y'|}{Y} \tag{11}$$

where N = The number of data points

RESULTS AND DISCUSSIONS

A. Regression results

Several sorption isotherms of cereal grains and oil seeds where selected to find the parameters of the GAB equation. These parameters and the values of P and S.E. are listed in Table 1. Almost all sorption isotherms have relatively small values of S.E., and almost all values of P are less than 5.

The residual plots of some sorption isotherms, including some food-stuffs for comparison, are shown in Figure 1-7. Because the GAB equation only has one independent variable (Aw), the residuals of the dependent variable (moisture content) are plotted against Aw except fishflour.

These residual plots show that high protein and oil materials (soybeans and fish flour) have residual plots with uniformly scattered data points. However, two regions of deviation of the residual points are found in high starch products.

Table 1. Estimated parameters and criteria for the GAB equation of cereal grains and seeds

		Parameters			Criteria	
Products	Temp. (°C)	Mo	K	C	S.E.	P
Red Beans	21.1	7.5231	0.8449	85.547	0.496	2.114
Guevaraguio	26.6	6.9178	0.8874	43.310	0.160	1.128
1973	32.2	6.5530	0.9292	48.264	0.122	0.955
	37.8	6.7608	0.9208	18.268	1.415	3.605
Corn Kernel	25.0	9.7569	0.6235	16.3156	0.153	2.595
Endosperm	25.0	10.1870	0.6386	21.2479	0.261	4.467
Shelef 1966						
Rice	10.0	10.7944	0.6254	28.1643	0.239	1.651
Califonia short	20.0	10.1848	0.6390	21.1141	0.261	2.062
1979	25.0	9.4453	0.6720	19.4670	0.191	1.468
	30.0	9.7540	0.6236	16.2810	0.153	1.435
	40.0	9.0560	0.6221	12.8950	0.157	1.969
Rice	10.0	10.9698	0.6430	15.2630	0.559	1.797
Austrila short	20.0	9.5820	0.6767	20.0950	0.501	2.825
1979	25.0	8.5489	0.7321	39.6826	0.195	0.994
	30.0	10.9970	0.9074	10.4060	0.172	1.112
	38.0	9.9778	0.6373	9.9230	0.177	1.349
Sorghum	15.6	10.534	0.6561	90.9250	0.728	5.476
Duggle	23.9	10.018	0.6680	57.2930	0.487	4.095
1972	26.7	10.553	0.6400	38.581	0.206	0.804
	40.6	9.269	0.6843	39.676	0.406	1.568
	48.9	9.999	0.6230	28.414	0.298	2.583
Soybeans	5.0	5.6230	0.8923	89.5405	0.395	2.335
Alam	15.0	5.4031	0.8936	40.6913	0.282	1.846
1972	25.0	4.9400	0.9298	48.7413	0.136	1.133
	35.0	4.6299	0.9410	29.5690	0.182	1.502
	45.0	4.4561	0.9359	17.0090	0.172	1.657
	55.0	4.0165	0.9542	13.3139	0.241	3.261
Wheat	25.0	8.6172	0.7470	25.8568	0.381	2.013
Becker	50.0	7.0076	0.7845	13.9072	0.256	1.552
1956						

Table 2. The GAB parameters of some foodstuffs

Products	Temp. (°C)	Mo	K	C
1. Roast ground coffee (Weisser) 1986	20	3.49	0.963	16.65
	25	3.22	0.993	15.87
	40	3.12	0.987	12.45
	60	2.74	1.029	8.59
	80	2.74	1.029	5.22
2. Durum wheat pasta (Andrieu, et al.) 1986	40	5.60	0.79	11.7
	50	6.20	0.85	21.1
	60	7.00	0.88	28.6
	70	11.30	1.28	36.3
	80	23.00	2.47	14.9
	90	26.00	3.12	12.6
3. Corn meal (Labuza, et al.) 1985	25	8.223	0.6879	15.680
	30	6.621	0.7857	19.293
	45	5.685	0.8496	15.965
	65	5.574	0.8231	8.431
4. Fish flour (Labuza, et al.) 1985	25	5.082	0.8537	6.647
	30	4.318	0.9030	5.417
	45	3.324	0.9932	6.621
	65	3.751	0.9458	3.087
5. Wide rice (Gencturk, et al.) 1987	10	10.317	0.6953	412.68
	24.5	7.377	0.7491	68.60
	38.0	7.046	0.7358	37.16
	43.5	6.946	0.7510	17.74

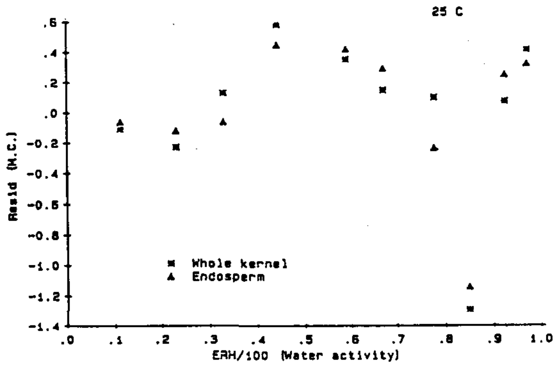

Fig. 1. Residual plots of the GAB equation for corn kernels

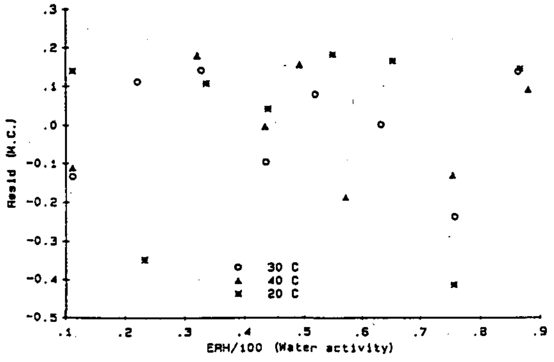

Fig. 2. Residual plots of the GAB equation for rice krnels.

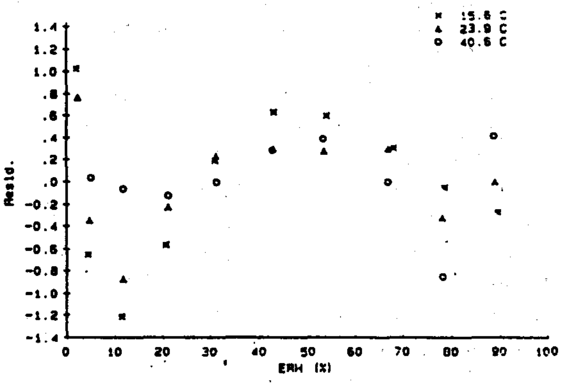

Fig. 3. Residual plots of the GAB equation for sorghum.

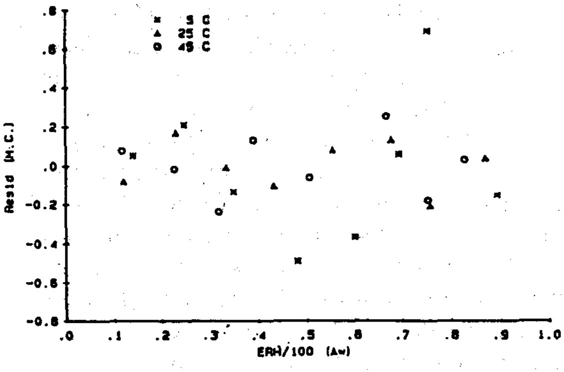

Fig. 6. Residual plots of the GAB equation for corn meal.

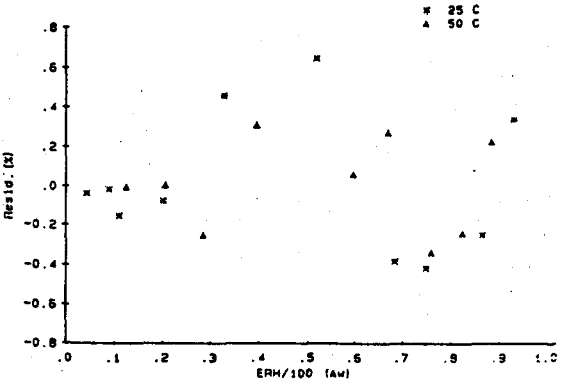

Fig. 4. Residual plots of the GAB equation for wheat.

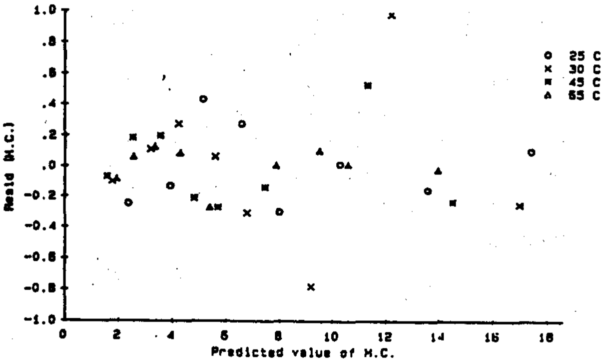

Fig. 7. Residual plots of the GAB equation for fish flour.

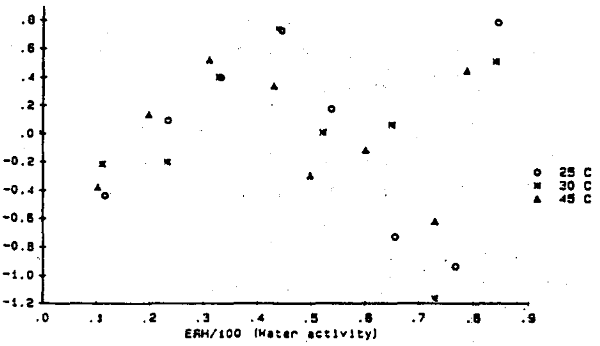

Fig. 5. Residual plots of the GAB equation for soybeans.

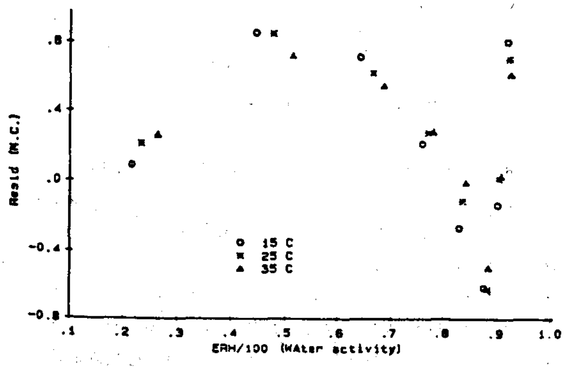

Fig. 8. Residual plots of special empirical equation for rapesseed (Candle).

One exists in the high RH range (0.7–0.8 Aw) for most residual plots, and 0.8–0.9 Aw for wheat. The other is in the low RH range but is not as significant as in the high RH range. These patterns can also be found for other starchy grains. The reason for this phenomenon may be explained by the fact that there are two sharp regions of the sigmoid curve for sorption isotherms of starchy grains; therefore the smooth GAB curve does not fit well in these regions. This lack of fit for starchy foods has been mentioned by Bizot (1984). Although the values of S.E. and P are low for this equation, the residual plots indicate its unsuitability for starchy products.

B. A particular case: Rapeseed

When computing the estimated parameters of the GAB equation for some sorption isotherms of rapeseed, it is very difficult to obtain convergence for the C value. There are two methods to find the estimated parameters in nonlinear regression models, neither the Gauss-Newton nor the steepest descent method could obtain the convergence value of C. The value for C always approached infinity, no matter what numerical techniques was used.

When the values of C is infinite, the GAB equation reduces to the following:

$$M = Mo^* \left(\frac{1}{1 - K^*Aw} \right) \qquad (12)$$

After rearranging the equation, it becomes a linear one,

$$\frac{1}{M} = A - B^*Aw \qquad (13)$$

Where A= 1/Mo and B= K/Mo

This reduced form of the GAB equations was used to fit those special sorption data, Table 3. The A and B parameters and R-square value are shown in Table 4.

High R-square values only indicate that a strong relationship exits between 1/M and Aw. As metnioned previously, they do not necessarily mean that GAB equation, complete from or reduced from, is not suitablefor these data because the former did not work, and the latter resulted in a definite pattern in the residual plots, Fig. 8.

Table 3. The relationship between moisture content and ERH of rapeseed

M.C. (%, d.b.)	Temperature (°C) 5.0	15.0	25.0	35.0
A. Tower rapeseed, adsorption				
4.60	27.2	30.4	34.0	37.7
6.38	47.8	50.0	52.8	55.2
9.41	70.4	70.8	72.4	72.9
12.99	80.9	81.9	82.2	84.1
20.92	91.0	91.7	91.8	92.0
B. Candle rapessed, adsorption				
4.16	–––	21.5	23.5	26.5
6.38	42.0	44.5	47.7	51.5
8.70	62.5	64.2	66.5	68.5
11.11	75.2	76.0	77.0	78.0
13.64	82.3	83.0	83.5	84.0
16.28	86.7	87.5	88.0	88.3
19.05	89.7	90.0	90.2	90.5
21.95	91.5	91.7	92.0	92.3
C. Candle rapeseed, adsorption				
6.38	46.5	48.7	50.5	52.5
8.70	62.0	66.0	66.5	68.7
11.11	76.0	76.5	77.3	78.3
13.64	82.3	83.0	83.5	84.0
16.28	86.7	87.5	88.0	88.3
19.05	89.7	90.0	90.2	90.5
21.95	91.5	91.7	92.0	92.3

Date source: Pixton and Warburton, 1977.

C. The temperature dependence of the GAB equation

There are two methods to consider the temperature effect for the sorption isotherms. One is to apply thermodynamic principles, for example, the Calusius-Clapeyron equation. The other method is to incorporate a temperature term into the sorption equations.

From the definition of parameters for the GAB equation (Van den Berg, 1984; 1985), C and K were also fit with an Arrhenius-type equation.

Table 4. The parameters and R-square value of some rapeseeds for an empirical equation: $1/M = A + B*Aw$

Products	Temp.	A	B	R^2
Tower Adsorption	5.0	0.2859	0.2599	0.9975
	15.0	0.2972	0.2712	0.9980
	25.0	0.3135	0.2888	0.9985
	35.0	0.3295	0.3049	0.9975
Candle Desorption	5.0	0.2517	0.2203	0.9941
	15.0	0.2868	0.2614	0.9892
	25.0	0.2971	0.2713	0.9937
	35.0	0.3121	0.2867	0.9969
Candle Adsorption	5.0	0.2724	0.2440	0.9965
	15.0	0.2819	0.2538	0.9969
	25.0	0.2916	0.2636	0.9969
	35.0	0.3035	0.2757	0.9960

$$Y = A*Exp(-B/Tabs) \tag{14}$$

There Tabs is the absolute temperature, (K)

This relationship can be applied for the Mo values (Iglesias and Chirife, 1984). Weisser (1985, 1986) incorporated this Arrhenius-type equation into three GAB parameters and developed a new temperature dependent, six parameter GAB(T) equation.

If Arrhenius-type relations for Mo, K, and C are not valid, Weisser's GAB(T) equation cannot be claimed as a temperature dependent equation with physical meaning. The F-ststistical technique can be used to evaluate the validity of these relationships.

Five sets of GAB parameters for cereal grains and seeds were selected over a wide temperature range. Parameters for several food materials taken from the literature are also included and are listed in Table 2. The results of F-tests are shown in Tables 5.

From the results, only the parameters from soybeans have a significant relation for an Arrhenius-type function with temperature. Some high protein and oil products, beans and fish flour, showed good agreement between sorption isotherm data and the GAB equation at each temperature. However, a statistically signicant relation with temperature did not exit for the parameters. Although the Mo values for Durm wheat pasta indicated a strong relation with temperature, the relation was in the wrong direction, that is, the Mo value increased as temperature increased. Sorption isotherms illustrate the application of the GAB(T) equation, however, no significant relation between K and temperature was found for these data.

Although the Arrhenius-type function has a physical meaning for three parameters, a "universal" temperature relation function was not found to incprorate into the GAB parameters in practical applications. A simple, accurate, and widely applicable temperature dependent GAB equation needs development. For example, the polynomial equation of temperature relation GAB equation of Durum wheat pasta (Andrieu, et al, 1986)

These equation are listed as following:

$$Mo = 23.134 - 0.843*T + 0.009929*T*\%, \quad R = 0.95$$

$$K = 3.756 - 0.1279*\% + 0.0013554*T*T, \quad R = 0.98$$

$$C = -100.2 + 4.023*T - 0.0311*T*T, \quad R = 0.74$$

D. The evaluation of the GAB equation for cereal grains ans seeds

A good fit for the GAB equation has been found for many foods and components. However, there are several reasons why this model does not meet the requirements of simplicity and accuracy necessary for agricultural products.

1. The expression of ERH as a function of moisture content is very complex.
2. The residual plots reveal that two regions of deviation exist for most starchy grains.
3. The equation only can express the relationship between Aw and temperature at fixed temperature. The Arrhenius-type function did not had the strong relation for GAB parameters and temperature. Although the polynomial equation have the better ability, it is difficult to incorporate temperature

Table 5. The results of the F-tests for the Arrhenius relationship between the GAB parameters and temperature.

Products	No. data	para.	A	B	F-value	results
Red beans	4	Mo	0.8899	–620.76	4.27	
		K	4.7080	502.15	10.05	
		C	9.6818E–10	–7411.6	10.13	
Rice (Calif.)	5	Mo	1.7913	–507.01	24.08	*
		K	0.5748	–30.2246	0.040	
		C	8.1648E–3	–2307.8	563.0	
Rice (Austr., short)	5	Mo	6.2789	–144.397	0.20	
		K	0.4363	–120.37	0.17	
		C	0.006645	–1642.02	0.41	
Sorghum	5	Mo	4.6542	–234.30	2.048	
		K	0.5291	–64.469	0.21	
		C	4.6876E–3	–2792.83	13.02	*
Soybeans	6	Mo	0.6521	–604.49	182.1	**
		K	0.7092	127.67	28.2	**
		C	6.0301E–$	–3292.8	48.0	**
Roast ground coffee (Weisser, 1986)	5	Mo	1.0947	–332.09	17.0	*
		K	1.2928	83.559	8.07	
		C	1.3958E–2	–2117.5	76.3	**
Durum wheat pasta (Andrieu et al., 1986)	6	Mo	1118550	3893.12	17.0	**
		K	27399	3347.5	31.9	**
		C	17.407	–30.796	0.001	
Corn meal (Labuza, et al., 1985)	4	Mo	0.4045	–868.317	5.69	
		K	2.6495	381.91	2.30	
		C	0.05533	1740.2	56.68	
Fish flour (Labuza, et al., 1985)		Mo	0.3720	–750.227	2.28	
		K	2.084	255.59	1.96	
		C	0.029	–1028.1	3.59	
Wide rice (Gencturk, et al.)	4	Mo	2.54E–3	–1033.3	11.4	
		K	1.3089	175.1	3.98	
		C	2.638E–10	–7914.0	62.0	

(Note) **: 99% significance, *: 95% significance.2

terms for three parameters. The model becomes even more complex involving more parameters.

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