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# Moisture adsorption isotherms of baru almond flours

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**ABSTRACT:** The whole meal (WBF) and partially defatted Baru almond (PDBF) flour have commercial and nutritional attractions. The study of the adsorption behavior is essential for correct storage. Thus, the objective was to determine the moisture adsorption isotherms for WBF and PDBF, at temperatures of 20, 30, and 35 °C, in the range of water activity from 0.070 to 0.975. The static gravimetric method was used. Fourteen models were adjusted to the experimental hygroscopic equilibrium humidity data. The GAB model better represented the moisture adsorption isotherms for WBF, while the BET model represented the PDBF. The curves of the two flours were classified as type III. Safe storage for WBF occurs at equilibrium humidity of 6.41, 7.12, and 7.49% d.b., and for PDBF at 9.04, 9.26, and 9.41% d.b., at the respective temperatures of 25, 30, and 35 °C.

**Key words:** BET model; *Dipetryx alata* Vogel; GAB model; moisture equilibrium; water sorption

# Isotermas de adsorção de umidade de farinhas de amêndoa de baru

**RESUMO:** As farinhas integrais (WBF) e parcialmente desengordurada das amêndoas de Baru (PDBF) possuem atrativos comerciais e nutricionais. O estudo do comportamento de adsorção é essencial para a correta armazenagem. Assim, objetivou-se determinar as isotermas de adsorção de umidade para WBF e PDBF, nas temperaturas de 20, 30 e 35 °C, no intervalo de atividade de água de 0,070 a 0,975. Utilizou-se o método gravimétrico estático. Quatorze modelos foram ajustados aos dados experimentais de umidade de equilíbrio higroscópico. O modelo de GAB representou melhor as isotermas de adsorção de umidade para WBF, enquanto o de BET, as de PDBF. As curvas das duas farinhas foram classificadas como tipo III. O armazenamento seguro para WBF ocorre em umidades de equilíbrio de 6,41, 7,12 e 7,49% b.s., e para PDBF em 9,04, 9,26 e 9,41% b.s., nas respectivas temperaturas de 25, 30 e 35 °C.

**Palavras-chave:** modelo BET; *Dipetryx alata* Vogel; modelo GAB; umidade de equilíbrio; sorção de água

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## Introduction

Baru (*Dipteryx alata* Vogel) is a tree belonging to the Fabaceae family found in the Brazilian Cerrado. The fruit produces a single seed, which has high levels of protein and oil, enhancing its consumption as roasted almonds, cakes, as well as the extraction of lipids (Paglarini et al., 2018). Baru oil has high social and commercial importance and is also exported. One of the byproducts of mechanical oil extraction is partially defatted Baru flour, with high potential to be included in a healthy human diet (Caetano et al., 2017).

Baru and it's processed foods, like most agricultural products, have a hygroscopic characteristic, that is, losing or gaining moisture with the environment, tending to equilibrium. The study of this behavior aims to maintain the quality of products, aiming to reduce the action of degrading agents. Knowledge of the sorption phenomenon is important to predict changes in water content, the point at which it increases and provides microbiological development, intense respiration, and warming (Oliveira et al., 2017).

An important factor influencing the sorption mechanism is the chemical constitution and structure of the food, such as the oil content. Therefore, experimentation is crucial to elucidate this process (Xu et al., 2019). In this sense, the determination of adsorption isotherms is important. These curves provide input for proper packaging selection, storage stability information, and maintenance of sensory properties. Several mathematical models are proposed in the literature to describe the adsorption isotherms; however, different products fit different models. Among the models and equations can be cited those proposed by Brunauer-Emmett-Teller (BET) and Guggenheim-Anderson-de-Boer (GAB) (Peleg, 2020). The hygroscopic behavior of many food and agricultural products has been explained by these two models (Bastıoğlu et al., 2017; Ahmed et al., 2018; Jung et al., 2018; Xu et al., 2019).

Some studies have addressed the hygroscopicity of Baru by-products, such as almonds and epicarp flour (Furtado et al., 2014; Oliveira et al., 2017; Resende et al., 2017). However, there is no approach to the sorption behavior of both the whole and partially defatted almond flours and the effect of oil content on it. This fact harms agro-extractive producers.

The objective of this study was to determine the moisture adsorption isotherms for the whole meal Baru almond (WBF) and partially degreased Baru (PDBF) flour at temperatures of 20, 30, and 35 °C, in the range of water activity from 0.070 to 0.975, as well as to determine the best mathematical model for the experimental data of hygroscopic equilibrium humidity.

## Materials and Methods

The almonds were collected from a native vegetation fragment of the Cerrado, in the municipality of Montes Claros, MG, Brazil. The experiment was conducted in laboratories of the Universidade Federal do Mato Grosso, Campus Rondonópolis, in the municipality of Rondonópolis, MT, Brazil.

The almond's superficial skins were previously removed and processed in a household blender, crushing about 100 g for 56 seconds (on average) at minimum speed. The flour was homogenized with the aid of a domestic sieve. Partial extraction of the oil was performed chemically, in an Oil and Grease Extractor by Immersion. Portions of approximately 50 g of WBF were immersed 1 hour and 30 minutes in Hexane solvent, at 100 °C having as a product PDBF.

After obtaining the flours, the lipid content of the WBF and PDBF was determined in duplicate, according to the methodology established by the Adolfo Lutz Institute (2008).

Moisture adsorption isotherms were determined for WBF and PDBF samples, previously oven-dried, at 105 ± 3 °C, until a constant mass, using the static gravimetric method at three temperatures (25, 30, and 35 °C). The test was conducted by saline containers described in Table 1 (providing different relative moistures) and the flour samples in triplicate were placed in a B.O.D. (Biochemical Oxygen Demand) oven.

The mass variation of the samples was periodically measured on an analytical balance until a constant weight was reached. Hygroscopic equilibrium humidity was determined by the Eq. 1:

$$M_e = \frac{W_m}{D_m} \cdot 100 \quad (1)$$

where:

$M_e$ - hygroscopic equilibrium moisture, % d.b.;
$W_m$ - water mass, g; and,
$D_m$ - dry mass, g.

Moisture Adsorption Isotherms curves were generated relating the equilibrium humidity to water activity. The experimental data were adjusted, using the SigmaPlot 14.0 software, to fourteen mathematical models described in Table 2.

**Table 1.** Water activity values (%) for saturated salt solutions under different temperature conditions.

| Salt | Temperature (°C) | | |
|---|---|---|---|
| | 25 | 30 | 35 |
| KOH | 8.0 | 7.5 | 7.0 |
| $MgCl_2 \cdot 6H_2O$ | 32.5 | 32.5 | 32.5 |
| $K_2CO_3$ | 43.0 | 42.0 | 41.0 |
| NaCl | 75.5 | 75.5 | 75.5 |
| $K_2SO_4$ | 97.5 | 96.5 | 96.0 |

**Table 2.** Mathematical models adjusted to the whole meal (WBF) and partially defatted Baru almond (PDBF) fluor hygroscopic equilibrium moisture data.

| Model name | Model | Eq. |
|---|---|---|
| BET | $M_e=\frac{(Xm\cdot c\cdot a_w)\cdot[1-(n+1)\cdot a_w{}^n+n\cdot a_w{}^{n+1}]}{(1-a_w)\cdot[1+(c-1)\cdot a_w-c\cdot a_w{}^{n+1}]}$ | (2) |
| Cavalcanti Mata | $M_e=[\ln(1-a_w)/(-a\cdot T^b)]^{\frac{1}{c}}$ | (3) |
| Chen Clayton | $M_e=[-1/(c\cdot T^d)]\cdot\ln[\ln(a_w)/(-a\cdot T^b)]$ | (4) |
| Chung Pfost | $M_e=a-b\cdot\ln[-(T+c)\cdot\ln(a_w)]$ | (5) |
| Copace | $M_e=\exp[a-(b\cdot T)+(c\cdot a_w)]$ | (6) |

Continues on the next page

Continuation of Table 2

| Model name | Model | Eq. |
|---|---|---|
| GAB | $M_e=\frac{(Xm\cdot c\cdot k\cdot a_w)}{(1-k\cdot a_w)\cdot(1-k\cdot a_w+c\cdot k\cdot a_w)}$ | (7) |
| Halsey | $M_e=(-c/\ln a_w)^{\frac{1}{n}}$ | (8) |
| Henderson | $M_e=[\ln(1-a_w)/(-a\cdot T^b)]^{\frac{1}{c}}$ | (9) |
| Modified Henderson | $M_e=\{\ln(1-a_w)/[-a\cdot(T+b)]\}^{\frac{1}{c}}$ | (10) |
| Oswin | $M_e=a\cdot[a_w/(1-a_w)]^b$ | (11) |
| Peleg | $M_e=(a\cdot a_w{}^b)+(c\text{-}a_w{}^d)$ | (12) |
| Sabbah | $M_e=a\cdot[a_w{}^b/T^c]$ | (13) |
| Sigma-Copace | $M_e=\exp\{a-(b\cdot T)+[c\cdot\exp(a_w)]\}$ | (14) |
| Smith | $M_e=a-(b\cdot T)+c\cdot\ln(1-a_w)$ | (15) |

Where: $a_w$ - water activity, dimensionless; T - temperature, °C; a, b, c, k, n, and C - adjustment parameters, dimensionless; $X_m$ - moisture in molecular monolayer, dimensionless.

The most appropriate fit was selected as a function of the determination coefficient ($R^2$), mean relative error (P) - Eq. 16, estimated mean error (SE) - Eq. 17, and chi-square ($\chi^2$) - Eq. 18:

$$P=\frac{100}{n}\sum_{i=1}^{n}\left(\frac{Me_{exp}-Me_{theo}}{Me_{exp}}\right) \quad (16)$$

$$SE=\sqrt{\frac{\sum_{i=1}^{n}\left(Me_{exp}-Me_{theo}\right)^2}{GLR}} \quad (17)$$

$$\chi^2=\frac{\sum_{i=1}^{n}\left(Me_{exp}-Me_{theo}\right)^2}{n-p} \quad (18)$$

where:

$Me_{exp}$ - experimental equilibrium moisture, % d.b.;
$Me_{theo}$ - equilibrium moisture predicted, % d.b.;
GLR - degrees of freedom of the model residue;
n - number of observed data; and,
p - number of model parameters.

## Results and Discussion

The lipid content test showed that WBF contained 45.60% oil in its content and PDBF 30.16%. The difference between the flours was approximately 15.00%. Experimental data for equilibrium humidity for WFB and PDFB are presented in Figure 1.

From the experimental data, it was observed that $M_e$ stabilized at higher $a_w$ contents and temperatures. For the same temperature condition and water activity, the PDBF reached the hygroscopic equilibrium in values higher than WBF. This fact occurs because PDBF is more hygroscopic than WBF, due to its lower oil content.

Similar results were found by Bo et al. (2017) when studying the sorption behavior of integral, partially defatted, and fully defatted pistachio flours. Pistachio flours with lower lipid contents reached the highest hygroscopic equilibrium $M_e$.

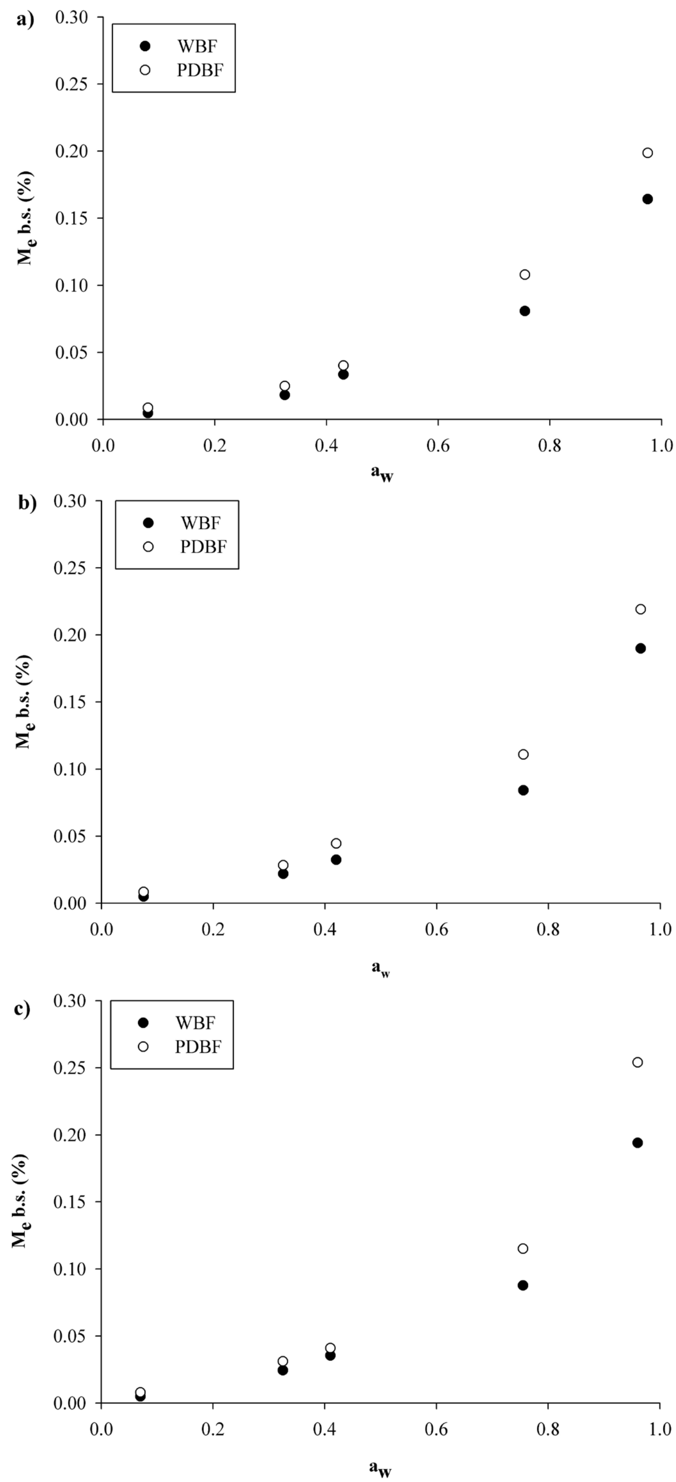



**Figure 1.** Experimental data graphic of $a_w$ vs. $M_e$ for WBF and PDBF at the temperatures 25 (A), 30 (B), and 35 °C (C).

The experimental data were adjusted to the fourteen models above, whose statistical criteria considered for selection are contained in Table 3.

Only Chen Clayton, Chung Pfost, Halsey, Oswin, and Sabbah models presented coefficients of determination below 99% for adjustments to WBF hygroscopic equilibrium data. To

**Table 3.** Estimated mean error (SE), mean relative error (P), determination coefficient ($R^2$), and chi-square ($\chi^2$) of the mathematical models' adjustment to the experimental data of $M_e$ for the wholemeal (WBF) and partially defatted Baru almond (PDBF) flour.

| Model | WBF | | | | PDBF | | | |
|---|---|---|---|---|---|---|---|---|
| | SE | P (%) | $R^2$ (%) | $\chi^2$ | SE | P (%) | $R^2$ (%) | $\chi^2$ |
| | | | | 25 °C | | | | |
| (2) | 0.0021 | 11.6933 | 99.96** | 0.0455 | 0.0007 | 2.1271 | 100.00** | 0.0049 |
| (3) | 0.4142 | 12.1847 | 99.85** | 0.1716 | 0.7306 | 7.7846 | 99.73** | 0.5338 |
| (4) | 1.3149 | 47.2604 | 99.25** | 1.7290 | 2.1506 | 49.7509 | 98.85** | 4.6249 |
| (5) | 0.9298 | 47.2574 | 99.25** | 0.8627 | 1.5207 | 49.7674 | 98.85** | 2.3089 |
| (6) | 0.5828 | 23.8507 | 99.71** | 0.3398 | 0.4987 | 13.7506 | 99.88** | 0.2491 |
| (7) | 0.0028 | 1.6392 | 99.86** | 0.1599 | 0.0021 | 6.0976 | 99.98** | 0.0442 |
| (8) | 1.8135 | 97.2494 | 95.74** | 6.5579 | 2.3971 | 72.5068 | 95.70** | 5.7461 |
| (9) | 0.0034 | 12.1789 | 99.85** | 0.1144 | 0.0060 | 7.7976 | 99.73** | 0.3559 |
| (10) | 0.4142 | 12.0773 | 99.85** | 0.1720 | 0.7306 | 7.6465 | 99.73** | 0.5354 |
| (11) | 1.1384 | 44.0687 | 97.69** | 2.4272 | 1.6691 | 41.2538 | 96.59** | 3.7181 |
| (12) | 0.0556 | 1.6202 | 100.00** | 0.0031 | 0.0951 | 1.4924 | 100.00** | 0.0090 |
| (13) | 1.5067 | 36.1384 | 98.04** | 2.2702 | 1.8076 | 36.4208 | 98.37** | 3.2676 |
| (14) | 0.8318 | 45.6622 | 99.40** | 0.6920 | 0.9577 | 31.0055 | 99.09** | 0.9175 |
| (15) | 0.7795 | 23.7626 | 99.48** | 0.6076 | 1.0292 | 14.6982 | 99.47** | 1.0593 |
| | | | | 30 °C | | | | |
| (2) | 0.0021 | 10.7071 | 99.96** | 0.3883 | 0.0033 | 4.5918 | 99.92** | 0.1127 |
| (3) | 0.2035 | 2.7437 | 99.96** | 0.0414 | 0.6688 | 6.9016 | 99.70** | 0.4474 |
| (4) | 1.9602 | 69.1783 | 98.29** | 3.8425 | 1.8255 | 41.6903 | 98.86** | 3.3323 |
| (5) | 1.3861 | 69.1337 | 98.29** | 1.9167 | 1.2908 | 41.6819 | 98.86** | 1.6634 |
| (6) | 0.6001 | 18.6317 | 99.68** | 0.3602 | 0.5262 | 14.6493 | 99.81** | 0.2772 |
| (7) | 0.0010 | 2.3010 | 99.99** | 0.0101 | 0.0027 | 7.6884 | 99.95** | 0.0724 |
| (8) | 1.5327 | 74.8656 | 96.86** | 2.3491 | 2.1803 | 63.6226 | 95.14** | 4.7539 |
| (9) | 0.1662 | 8.4574 | 99.96** | 0.6528 | 0.5461 | 9.3635 | 99.70** | 0.5861 |
| (10) | 0.2035 | 2.7647 | 99.96** | 0.0414 | 0.6688 | 6.8763 | 99.70** | 0.4474 |
| (11) | 1.0695 | 46.2412 | 98.47** | 1.1438 | 1.6214 | 41.9593 | 97.31** | 2.6289 |
| (12) | 0.1693 | 4.9401 | 99.99** | 0.0287 | 0.4144 | 6.5676 | 99.94** | 0.1717 |
| (13) | 1.5439 | 39.4883 | 97.88** | 2.3838 | 1.5022 | 30.5688 | 98.46** | 2.2566 |
| (14) | 0.6828 | 38.0537 | 99.59** | 0.4662 | 1.1161 | 35.3752 | 99.15** | 1.2462 |
| (15) | 0.2719 | 5.2317 | 99.93** | 0.0740 | 1.0563 | 17.3303 | 99.24** | 1.1159 |
| | | | | 35 °C | | | | |
| (2) | 0.0047 | 11.6879 | 99.74** | 0.2236 | 0.0036 | 9.0530 | 99.79** | 0.1289 |
| (3) | 0.5084 | 10.6241 | 99.69** | 0.2586 | 0.8969 | 11.6902 | 99.34** | 0.8045 |
| (4) | 1.6006 | 71.3286 | 98.48** | 3.0532 | 2.0052 | 42.2431 | 98.36** | 4.0726 |
| (5) | 1.1318 | 61.9664 | 98.48** | 1.2780 | 1.4179 | 45.3090 | 98.36** | 2.0081 |
| (6) | 0.6059 | 24.0456 | 99.56** | 0.3671 | 0.4728 | 14.6922 | 99.82** | 0.2236 |
| (7) | 0.0039 | 9.0557 | 99.81** | 0.1574 | 0.0036 | 12.0900 | 99.89** | 0.1330 |
| (8) | 1.5580 | 79.0012 | 95.67** | 2.4274 | 2.1721 | 60.3140 | 94.22** | 4.7179 |
| (9) | 0.0041 | 11.5842 | 99.69** | 0.2783 | 0.0073 | 12.6245 | 99.34** | 0.6886 |
| (10) | 0.5084 | 10.5793 | 99.69** | 0.2593 | 0.8969 | 11.7581 | 99.34** | 0.8055 |
| (11) | 1.3425 | 68.9664 | 97.67** | 1.6143 | 1.8205 | 48.3106 | 97.52** | 3.1672 |
| (12) | 0.5065 | 17.1238 | 99.85** | 0.2566 | 0.4623 | 6.1154 | 99.91** | 0.2137 |
| (13) | 1.2366 | 32.2612 | 98.18** | 1.5291 | 1.1155 | 26.8622 | 98.98** | 1.2444 |
| (14) | 0.9030 | 49.2918 | 99.03** | 0.8157 | 1.2186 | 34.4671 | 98.79** | 1.4851 |
| (15) | 0.6542 | 18.2887 | 99.49** | 0.4280 | 1.2087 | 15.8947 | 98.81** | 1.4610 |

** Significant at 1% probability by the t-test.

these models are added those of Sigma-Copace and Smith in the PDBF sorption study.

According to Barati et al. (2016) lower chi-square values are sought in mathematical modeling. Thus, the smaller magnitudes of $\chi^2$ were found for Cavalcanti Mata, GAB, Modified Henderson and Peleg models in WBF adjustments, while BET, GAB, and Peleg models were better for PDBF.

Regarding the statistical criterion of SE, only the BET and GAB models were suitable at all temperatures, for both WBF and PDBF. According to Draper and Smith (1998), the ability of a model to accurately predict a physical process is inversely proportional to the value of the estimated mean error (SE).

Only the GAB model presented P values below 10%, considering the three studied temperatures for WBF. In turn, this condition was observed for PDBF in the BET and Peleg models. According to Mohapatra & Rao (2005) models with a mean relative error greater than 10% are inappropriate to predict the physical phenomenon. It was also noted that the P values were higher as there was an increase in temperature, as well as the partially defatted flour presented a higher number of models with satisfactory P.

Therefore, according to the statistical criteria considered, the models selected for WBF and PDBF adjustments were GAB and BET, respectively. The selection of different flour models shows that the difference between the chemical constituents promotes different adjustments. Such a situation was found by Bo et al. (2017) in the study of pistachio flour, in which for the integral fat, the Smith model was better adjusted; however, for partially and totally defatted flour the Halsey model was selected. Table 3 shows the adjustment parameters for the selected models to predict moisture adsorption of Baru flour.

The range of moisture at the molecular monolayer was identified at 3.85 to 4.04% (d.b.) for WBF and 3.13 to 3.55% (d.b.) for PDBF. These values were smaller than raw and extruded sorghum flours, which contained 6.90 and 6.70% (d.b.) water content at $X_m$, respectively at 25 °C, estimated from the GAB model (Galdeano et al, 2018).

By comparing the present study and Galdeano et al. (2018), note that the estimated molecular monolayer moisture values were higher for the WBF (estimated by GAB model) when compared to PDBF at all temperatures. This fact can be explained by the higher oil content of the whole meal flour. As $X_m$ is a deeper layer adsorption water, fatty acid molecules act as a barrier to water movement due to its hydrophobic characteristic.

In addition to the oil content factor, the parameter $X_m$ may also have been influenced by the estimation by different models. Ahmed et al. (2018) studying the adsorption isotherms of wheat, rice and corn flours found that the GAB model provided higher values for moisture in the molecular monolayer than that of BET. However, this fact will denounce the product characteristics. For example, Choi & Lee (2018) noted the opposite values for the $X_m$ estimated by GAB and BET model, in which the calculated monolayer moisture was smaller for the first model.

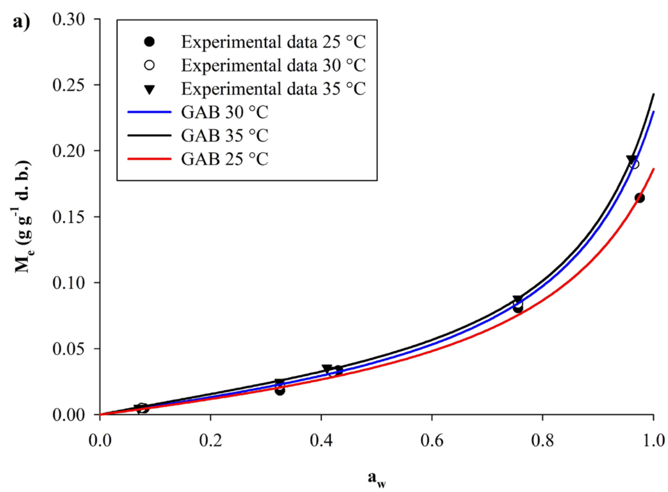
It is also observed that $X_m$ tended to decrease with increasing temperature in both flours. This behavior was verified by Alves et al. (2015) in the study of the desorption isotherms of green bell peppers. It was noted that the other parameters for both WBF and PDBF were directly proportional to temperature.

The GAB model was also selected to describe the whole grain sorption phenomenon of black pepper (Yogendrarajah et al., 2015). The same model was well-adjusted to moisture adsorption isotherms of flour of three banana and cassava varieties (Brou et al., 2018).

The moisture adsorption isotherms curves drawn by the GAB models for WBF and the BET model for PDBF are presented in Figure 2.

According to Figure 2, the isotherm curves can be classified as type III, or Flory-Huggins, according to Sing et al. (1985). According to the authors, these isotherms are of convex characteristic, not exhibiting an inflection, common in sigmoid isotherms, called point B. The absence of this deviation can be explained by the weak adsorbent-adsorbate interaction, which is common in hydrophobic materials, as in the case of these flours, due to the high oil content.

Another feature associated with the absence of point B, reported by Sing et al. (1985) are the low values of the constant C (<20), in the case of the BET model, which is found for PDBF. In these types of isotherms, the moisture adsorption



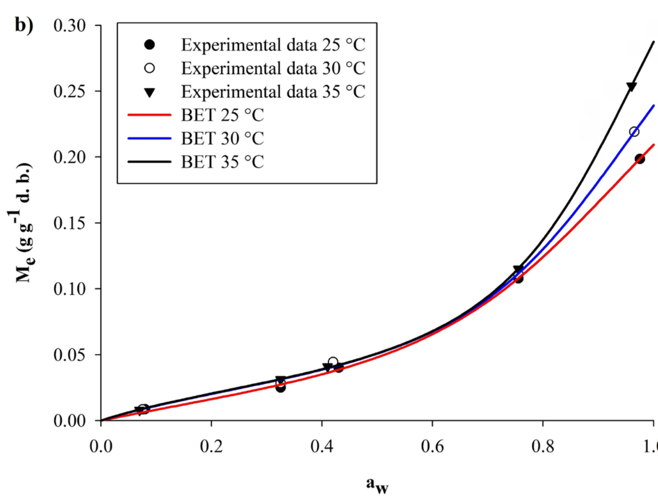




**Figure 2.** Moisture adsorption isotherms curves for the whole meal - WBF (A) and partially defatted Baru (PDFB) flour (B).

**Table 4.** Adjustment parameters for the mathematical fit of the GAB model for the whole meal (WBF) and the BET model for partially defatted Baru (PDBF) fluor.

| Temperature (°C) | Parameters of GAB model (WBF) | | | Parameters of BET model (PDBF) | | |
|---|---|---|---|---|---|---|
| | $X_m$ | C | k | $X_m$ | n | C |
| 25 | 0.0404** | 1.6815** | 0.8096** | 0.0355** | 11.9127** | 2.5456* |
| 30 | 0.0401** | 1.8789 * | 0.8412** | 0.0314** | 14.4921** | 4.2548* |
| 35 | 0.0385** | 2.4713 * | 0.8521** | 0.0313** | 17.6324** | 4.4449* |

** Significant at 1% probability by the t-test; * Significant at 5% probability by the t-test.

is low up to 0.8 of $a_w$, and after this value increases until $a_w$ = 1. Li et al. (2019) found type III moisture adsorption isotherms for Sultana raisin. Bastıoğlu et al. (2017) determined for microencapsulated extra virgin olive oil powder type III sorption isotherms, adjusted to the GAB model.

The temperatures used to evaluate the isotherms were selected in order to provide the experiment with conditions closer to commercialization, in which the products are exposed. It is noted that there is a narrow range of difference between the curves for each temperature, presenting a greater degree after 0.75 of water activity.

Oliveira et al. (2017) considered the minimum water activity to initiate fungal growth at 0.70 in the study of Baru fruit isotherms. Therefore, considering the same $a_w$ point, for safe storage for WBF, it is recommended that the water content (% d.b.) be lower than 6.41, 7.12, and 7.49, respectively, at temperatures 25, 30, and 35 °C. For PDBF, storage at equilibrium humidity of 9.04, 9.26, and 9.41% d.b. is ensured at the three studied temperatures, respectively.

## Conclusions

The best fit models were GAB for the whole meal flour and BET for partially degreased Baru flour.

Adsorption isotherms of Baru almond flour can be classified as type III.

For storage of the whole meal flour at a water content of 6.41, 7.12, and 7.49% d.b., and for partially defatted flour with 9.04, 9.26, and 9.41% d.b., respectively temperatures of 25, 30, and 35 ° C are recommended.

## Compliance with Ethical Standards

**Author contributions:** Conceptualization: NMCA, TAAS; Formal analysis: TAAS, NBCG; Funding acquisition: NMCA, TAAS; Investigation: TAAS, NBCG, IDS; Methodology: TAAS, NBCG; Project Administration: NMCA, TAAS; Supervision: NMCA; Validation: NMCA; Visualization: NMCA, TAAS, NBCG, IDS; Writing – original draft: TAAS, NBCG; Writing – review & editing: NMCA, TAAS, IDS..

**Conflict of interest:** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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