

Sorption isotherm modelling of dried tomatoes

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Abstract: The sorption isotherm (SI) of dried tomatoes was studied at three different temperatures, 15, 25, and 35 °C, using a static gravimetric method. The modified forms of the Guggenheim-Anderson-de Boer (mGAB), Halsey (mHAL), Henderson (mHEN), and Oswin (mOSW) models that incorporate the temperature term in their equation were selected and used to describe the experimental data of dried tomatoes. The mGAB model best described the SI of dried tomato samples at individual temperatures, having the highest coefficient of determination (R^2) and the lowest sum of squares of errors (SSE), the root mean square error (RMSE), and the corrected Akaike information criterion values (AIC_c). However, based on the statistical indices, three other tested models outperformed the mGAB model in describing the multi-temperature estimation to differentiate the temperature effect. The mOSW and mHAL models were superior in this case.

Keywords: Halsey model; modified GAB model; one-step SI modelling; Oswin model; temperature effect

Tomatoes (*Solanum lycopersicum* L.) are highly cultivated vegetables that provide a wide range of nutrients and many health benefits for humans. They are high in phytochemical carotenoids (lycopene), potassium, and vitamins C and E (Adebawale et al. 2022). In human diets, tomatoes are the main source of lycopene, a thermolabile carotenoid with effective antioxidant activity (Telis and Sobral 2002; Durigon et al. 2016). Tomatoes, especially ripe, are not suitable for long-term storage after harvest because they are very susceptible to rot. Consequently, drying is one of the most suitable processes for producing a finished product with a significantly reduced moisture content, thus preventing microbial degradation (Labuza and Altunakar 2007).

It is helpful to characterise the sorption properties and provide supporting knowledge to improve the shelf

life of dried food products. Knowledge of sorption isotherms (SI) is particularly important to identify optimal food storage conditions (Caballero-Cerón et al. 2015). They represent the relationship between the equilibrium moisture content and the water activity (a_w) in food or other materials at a constant temperature (Hartley et al. 2001). The water content or water activity as the variables of SI or intrinsic food factors are a good measure of various extensive undesirable effects on several aspects of food material, referring to the microbiological quality of food materials, different chemical reactions in foods, and specific physical attributes of foods (Fu and Labuza 1993). Furthermore, many of these aspects are non-linearly related to water content. At the same time, the relationship to a_w is mostly linear, favouring this intrinsic food environmental factor in predicting food sta-

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bility. For example, by decreasing $a_w < 0.6$, the growth of microorganisms can be eliminated, and by further dehydration of food products ($a_w < 0.3$), the rate of chemical reactions such as enzyme and non-enzymatic reactions also decreases substantially; however, a further decrease in the water activity of food material with a higher fat content can lead to an increase in the rate of lipid oxidation (Fontana 2000). Additionally, reducing the water activity of food materials to an extremely low level can also affect their physical properties and make them unacceptable to consumers. Therefore, studying sorption isotherms of various food materials is a valuable tool for improving their storage conditions and, consequently, their shelf life.

Although Mariem and Mabrouk (2015) focused on SIs of fresh tomato slices, there still appears to be a lack of reliable information on the moisture content and water activity relations of dried foods from developing countries. Dried tomatoes are characterised by a high specific surface associated with the risk of water absorption when entering contact with the surrounding air with higher relative humidity when a drop in temperature can reach a dew point. Thus, knowledge of sorption isotherm conditions in relation to storage conditions is crucial, as even a slight alteration in air-state conditions can lead to a significant change in water activity or moisture content. As a result of a_w value increase in a product, chemical, physical, enzymatic reactions and microbial proliferation can lead to food deterioration or health risks to consumers. Therefore, this study aims to provide sorption isotherms of traditional sun-dried tomatoes dried at high altitudes, low pressure, and relative humidity of the surrounding atmosphere, for example, in North-Central Afghanistan (Kabul). We aimed to model the sorption isotherms using modified models with incorporated temperature terms and evaluated their suitability in the range of temperatures of 15 to 35 °C which are typical for storage conditions in the above-mentioned region from June to October (Climates to travel 2024).

MATERIAL AND METHODS

Samples. The samples used in this research were traditionally sun-dried tomatoes originating in Afghanistan (personal information). The 3 kg of dried tomatoes shredded were obtained from local markets in Afghanistan in dehydrated form and transferred to the Slovak University of Technology in Bratislava (STU), where the experimental part of the research was carried out. Samples were kept inside a laboratory vacuum bag, in the dark, and at ambient laboratory temperature.

Method and measuring equipment. The adsorption and desorption isotherms of the dried tomato samples were evaluated according to Nayab et al. (2023) using a static gravimetric method at 15, 25, and 35 °C (± 0.5 °C, in all cases). In the experiments, saturated salt solutions that are used as water activity reference standards with known and constant water activity levels (Fontana Jr. 2007) were used. A Novasina LabMaster-aw instrument (Novasina, Switzerland) was used to check the a_w values of the working solutions periodically.

In detail, three to four series of saturated salt solutions with different water activity levels were prepared in jars with sealed lids before experiments. Approximately 2 g of the dried tomato sub-samples were cut to 3×3 mm weighed (± 0.0001), and placed in sieves inside the jars so as not to fall inside the saturated salt solution or inhibit the closure of the jars. They were then stored in thermostats (± 0.3 °C) until the samples reached a humidity equilibrium that usually lasted a week. To prevent moulding, toluene pots were placed inside the jars with saturated high a_w (Wolf et al. 1985).

The water content of the samples in equilibrium was immediately determined with a moisture analyser (MA X2.A; RADWAG, Poland) and recalculated to dry matter. Figure 1 represents an illustration of the experimental jar of this investigation.



Figure 1. Schematic diagram of the experimental setting (Nayab et al. 2023)

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Mathematical SI models and their performance. Modified forms of mathematical models, namely the Guggenheim-Anderson-de Boer (mGAB), Halsety (mHAL), Henderson (mHEN), and Oswin (mOSW) models that incorporate the temperature term in their equations, were applied to describe SI of dried tomatoes (Equations 1–4; Aviara 2020; Jolkili et al. 2020). Numerous researchers have reported these models for their ability to describe SI accurately (Peleg 2020):

$$M_w = \frac{AB \frac{C}{T} a_w}{\left(1 - Ba_w\right) \left(1 - Ba_w + B \frac{C}{T} a_w\right)} \quad (1)$$

mHAL model

$$M_w = \left(\frac{-e^{A+BT}}{\ln a_w} \right)^{\frac{1}{C}} \quad (2)$$

mHEN model

$$M_w = \left[\frac{-\ln(1 - a_w)}{A + BT} \right]^{\frac{1}{C}} \quad (3)$$

mOSW model

$$M_w = \left(A + BT \right) \left(\frac{a_w}{1 - a_w} \right)^{\frac{1}{C}} \quad (4)$$

where: M_w – equilibrium moisture content (EMC; g water per g dry matter); a_w – water activity; T – absolute temperature (K).

Non-linear regression analysis was used for the model parameters optimisation. The parameter A of the mGAB model is identical to the original M_0 and represents the monolayer moisture content at which the water is attached to the material surface. The constants B and C (originally reported as K and C) are related to the energies between the first and further layers of adsorbed water molecules (Rao 2016).

The statistical performance of the mathematical models adapted to the experimental adsorption and desorption data was first individually evaluated at each temperature (15, 25, 35 °C). Then multi-temperature parameter estimation was applied for all sorption isotherms to evaluate the temperature effect and used for predicting equilibrium moisture content (EMC) at given a_w within the range of 15 to 35 °C. The ran-

dom M_w residuals were obtained between the experimental and calculated values for each observation point (r_i). The following statistical indices, such as the sum of squares of errors (SSE), the sum of squares total (SST), the coefficient of determination (R^2), and the root mean square error (RMSE), were used to evaluate the goodness of fit of the models:

$$SSE = \sum_{i=1}^n \left(y_i^{\text{exp}} - y_i^{\text{cal}} \right)^2 \quad (5)$$

$$R^2 = 1 - \frac{SSE}{SST} \quad (6)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^n \left(x_i^{\text{exp}} - x_i^{\text{cal}} \right)^2}{n - p}} \quad (7)$$

where: y_i^{exp} and y_i^{cal} – observed and predicted values (% of g water per g dry basis), respectively; n – total number of data points; p – number of parameters.

The corrected Akaike information criterion (AIC_c) was used to compare the mathematical models.

$$AIC_c = n \ln \frac{SSE}{n} + 2(p + 1) + \frac{2(p + 1)(p + 2)}{n - p - 2} \quad (8)$$

The model with the smallest AIC_c value is more likely to be correct (Motulsky and Christopoulos 2003).

Their individual 95% confidence interval tested the statistical significance and the precision of the non-linear parameter estimates. Commercial process engineering software Athena Visual Workbench (version 8.3) and Microsoft Excel (version Microsoft 365) were applied to evaluate parameters and calculate the statistical indices mentioned above, respectively.

RESULTS AND DISCUSSIONS

Sorption isotherm curves at temperatures 15, 25, and 35 °C. The hysteresis between the adsorption and desorption values was not evident at lower water activities (< 0.8). In contrast, at higher levels of water activity (> 0.8), there may be a slight hysteresis between the adsorption and desorption curves at the three temperatures, 15, 25, and 35 °C (± 0.5 °C, in all cases). The experimental data on SI of dried tomatoes at 15, 25, and 35 °C and their fitted values by the mGAB model are presented in Figure 2.

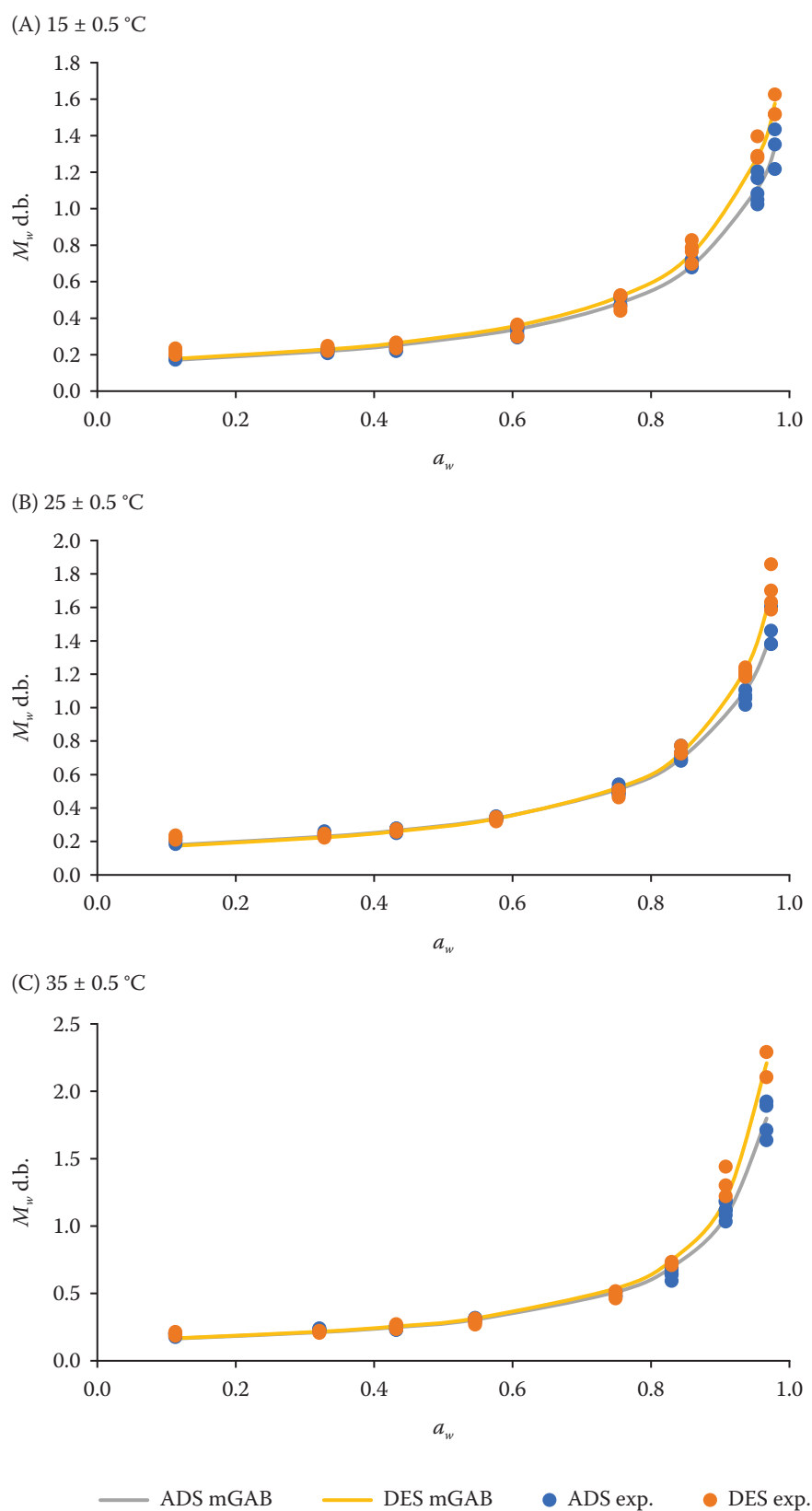


Figure 2. Adsorption and desorption isotherms of dried tomatoes at (A) 15 °C, (B) 25 °C, and (C) 35 °C

M_w d.b. – moisture content per dry basis; ADS – adsorption; DES – desorption; mGAB – Guggenheim-Anderson-de Boer model; exp. – experimental values

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Comparison of different models to describe the experimental data at individual temperatures.

The mGAB model (Equation 1) could fit adsorption and desorption isotherms at all individual temperatures with the highest coefficient of determination, > 0.997 . Similarly, Vishwakarma et al. (2011) and Yogendrarajah et al. (2015) considered the GAB model the best. Likewise, other applied mathematical models also performed quite well in predicting adsorption and desorption values at individual temperatures (15, 25, 35 °C) with determination coefficients of > 0.977 and > 0.971 for mHAL (Equation 2), > 0.970 and > 0.971 for mHEN (Equation 3), and > 0.984 and > 0.975 for mOSW (Equation 4) for adsorption and desorption, respectively. Comparable coefficients of determination were found for the GAB and Halsey models by Demarchi et al. (2013) in tomato pulp formulations at 20 and 40 °C ($R^2 > 0.988$ and $R^2 > 0.998$; respectively). However, the Peleg model was evaluated as the best of 12 empirical models used by Hadrich et al. (2016) for the adsorption and desorption isotherms of Tunisian tomatoes at higher temperatures of 45 and 55 °C (> 0.993 and > 0.999 , respectively).

Tables 1 and 2 summarise the parameters of the modified models with their 95% confidence limit for adsorption and desorption isotherm curves at all tested temperatures. The 95% confidence level was not determined for parameter C of the mGAB model. However, the other two parameters of the GAB model are the most influential parameters on the shelf life of selected food products, as reported in the literature (Escobe-

do-Avellaneda et al. 2012). The monolayer moisture content for the dried tomato samples was in the range of 0.147 ± 0.014 to 0.161 ± 0.013 , calculated by the mGAB model. Monolayer sorption values of similar food commodities have been reported for tomato slices in the range of 0.119 to 0.205 (Akanbi et al. 2006) and 0.145 to 0.229 (Adebawale et al. 2022) and for spray-dried tomato pulp in the range of 0.200 to 0.213 (Goula et al. 2008).

When all calculated statistical indices are compared (Table 3), we can infer that mGAB is the best model for fitting adsorption and desorption values (desorption results are not shown) at any individual temperature. In all cases, the values of the R^2 and AIC_c criterion were > 0.997 and < -33.92 for the mGAB model and < 0.992 and > -31.16 for all other tested models, respectively, clearly shows the superiority of the mGAB model compared to the mHAL, mHEN, and mOSW models to predict the adsorption and desorption isotherms of dried tomato samples at individual temperatures of 15, 25, and 35 °C. Regardless of the model used, the correlations between the calculated and experimental data points (n) were characterised by R values much higher than R_{crit} values in the range of 0.381 to 0.288 for $\alpha = 0.05$ and n between 25–40 (Štěpánek 1972). It also provides evidence that the models described the experimental data well.

The residuals M_w d.b. for mGAB dried tomato adsorption and desorption isotherms at 15, 25, and 35 °C are presented in Figure 3.

Table 1. Parameters of the Guggenheim-Anderson-de Boer (mGAB), Halsey (mHAL), Henderson (mHEN), and Oswin (mOSW) models for dried tomato adsorption isotherms at 15, 25, and 35 °C with the 95% confidence limits

Parameters	mGAB	mHAL	mHEN	mOSW
15 °C				
A	0.154 ± 0.007	-14.564 ± 0.213	-0.036 ± 0.003	-1.268 ± 2.476
B	0.904 ± 0.006	0.040 ± 0.001	-367.710 ± 5.830	0.006 ± 0.0001
C	$1.717 \times 10^{5*}$	2.380 ± 0.436	1.100 ± 0.197	2.630 ± 0.409
25 °C				
A	0.161 ± 0.013	-14.640 ± 0.125	-0.036 ± 0.004	-1.309 ± 0.044
B	0.913 ± 0.011	0.040 ± 0.0004	-367.710 ± 6.110	0.006 ± 0.00005
C	$3.004 \times 10^{5*}$	2.185 ± 0.253	1.049 ± 0.235	2.424 ± 0.261
35 °C				
A	0.147 ± 0.014	-14.680 ± 0.127	-0.037 ± 0.004	-1.398 ± 0.063
B	0.949 ± 0.010	0.040 ± 0.0003	-367.710 ± 4.630	0.006 ± 0.0001
C	$6.640 \times 10^{5*}$	1.728 ± 0.214	0.780 ± 0.194	1.893 ± 0.257

* The 95% confidence limit was not determined

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Table 2. Parameters of the Guggenheim-Anderson-de Boer (mGAB), Halsey (mHAL), Henderson (mHEN), and Oswin (mOSW) models for the desorption isotherms of dried tomatoes at 15, 25, and 35 °C with the 95% confidence limits

Parameters	mGAB	mHAL	mHEN	mOSW
15 °C				
A	0.159 ± 0.016	−14.253 ± 0.244	−0.031 ± 0.003	−1.250 ± 0.090
B	0.918 ± 0.012	0.040 ± 0.001	−367.710 ± 6.840	0.006 ± 0.0001
C	2.103 × 10 ^{5*}	2.240 ± 0.472	0.993 ± 0.228	2.450 ± 0.490
25 °C				
A	0.156 ± 0.012	−14.424 ± 0.135	−0.033 ± 0.003	−1.314 ± 0.061
B	0.933 ± 0.009	0.040 ± 0.0004	−367.710 ± 5.830	0.006 ± 0.00006
C	3.680 × 10 ^{5*}	1.981 ± 0.248	0.902 ± 0.220	2.176 ± 0.288
35 °C				
A	0.149 ± 0.014	−14.477 ± 0.129	−0.034 ± 0.003	−1.405 ± 0.066
B	0.964 ± 0.007	0.040 ± 0.0003	−367.710 ± 3.620	0.006 ± 0.0001
C	6.640 × 10 ^{5*}	1.549 ± 0.177	0.664 ± 0.149	1.680 ± 0.213

* The 95% confidence limit was not determined

Sorption isotherm modelling using a multi-temperature approach. Next, a multi-temperature approach was applied to observe dried tomato samples' adsorption and desorption curves. The mGAB model

was the best in the individual estimation of adsorption/desorption isotherms in a single temperature approach, in terms of having higher coefficient of determination values and lower SSE, RMSE, and AIC_c val-

Table 3. Statistical indices for the Guggenheim-Anderson-de Boer (mGAB), Halsey (mHAL), Henderson (mHEN), and Oswin (mOSW) adsorption isotherm modelling of dried tomatoes at 15, 25, and 35 °C

Models	SSE	R^2	RMSE	AIC_c
15 °C*				
mGAB	0.001	0.999	0.013	−52.15
mHAL	0.031	0.977	0.079	−23.08
mHEN	0.022	0.984	0.066	−25.85
mOSW	0.021	0.984	0.065	−26.18
25 °C**				
mGAB	0.003	0.998	0.026	−41.05
mHAL	0.014	0.990	0.053	−29.42
mHEN	0.038	0.974	0.087	−21.56
mOSW	0.011	0.992	0.048	−31.16
35 °C***				
mGAB	0.006	0.997	0.036	−35.67
mHAL	0.024	0.989	0.069	−25.28
mHEN	0.067	0.970	0.115	−16.98
mOSW	0.026	0.988	0.072	−24.48

* Number of data points $n = 32$, ** $n = 41$, *** $n = 38$; R^2 – coefficient of determination; SSE – sum of squares of errors; RMSE – root mean square error; AIC_c – corrected Akaike information criterion values

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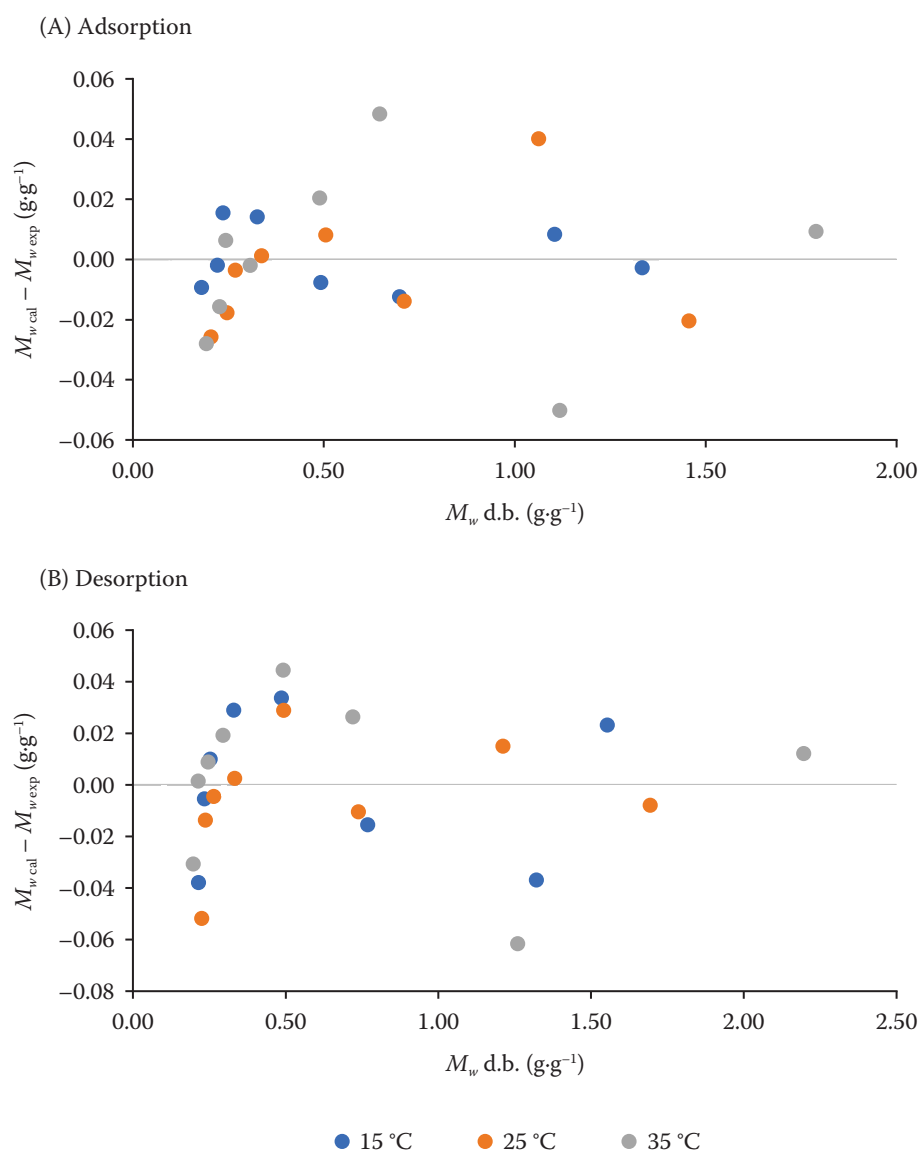


Figure 3. A random plot of the moisture content per dry basis (M_w d.b.) residuals for the mGAB dried tomato sorption isotherms – (A) adsorption and (B) desorption

mGAB – Guggenheim-Anderson-de Boer model; $M_{w,cal}$ – moisture content calculated; $M_{w,exp}$ – moisture content experimental

ues than the other selected models. However, other applied models outperformed the mGAB model in the case of multi-temperature estimation. The determination coefficient of the mGAB model, in this case, was calculated as 0.947 and 0.935 for the adsorption and desorption curves, respectively, which is lower than the three other models tested, which are > 0.967 , > 0.961 , and > 0.967 for the mHAL, mHEN, and mOSW models, respectively. Figure 4 shows the modelling of the multi-temperature adsorption isotherms of dried tomatoes using the mOSW and mHAL models, which, in this case, had the highest determination coefficients. The Oswin model has also been reported to be a su-

prior sorption isotherm model in addition to the GAB model in the literature (Qadri et al. 2022). Similarly, the value of the AIC_c criterion for the mGAB model was -98.09 , which was higher than the values of the same criterion calculated for the mHAL, mHEN, and mOSW models as -112.80 , -107.04 , and -114.81 , respectively. Table 4 represents the parameters of the selected modified models, and Table 5 summarises the statistical indices related to the multi-temperature estimation to describe the SI of dried tomatoes.

According to Akanbi et al. (2006), which evaluated the GAB and Oswin models as the best model to describe the isotherm of dehydrated tomato

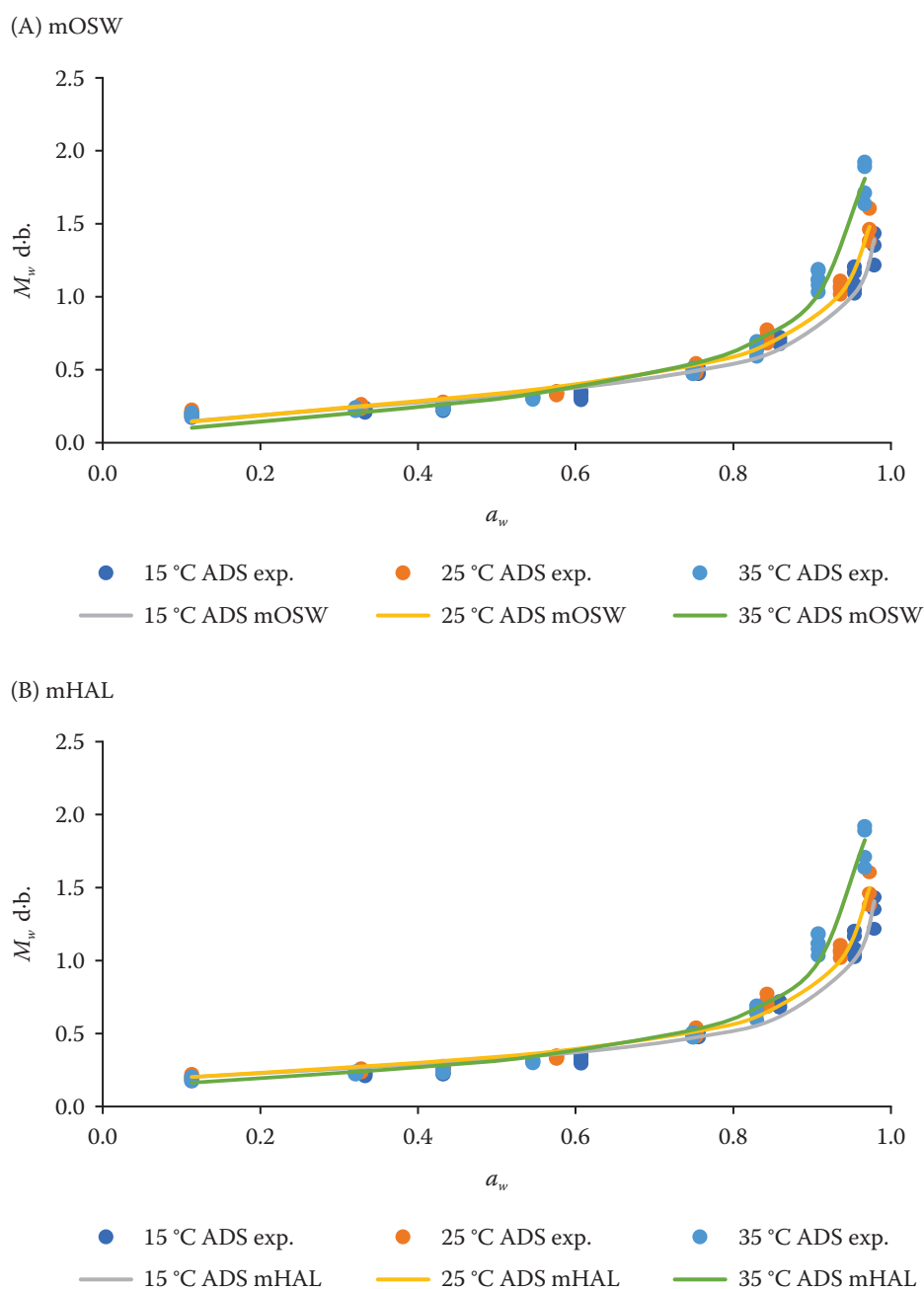


Figure 4. Multi-temperature estimation to adsorption isotherms of dried tomatoes for (A) mOSW model and (B) mHAL model

mOSW – Oswin model; mHAL – Halsey model; M_w d.b. – moisture content per dry basis; ADS – adsorption; exp. – experimental values

adsorption at 25 °C, our results and the statistical indexes in Table 5 also support their experience. Recently, Loumani et al. (2020) have shown for dried tomato desorption isotherms and Adebawale et al. (2022) for adsorption isotherms of dried tomato slices that the GAB model was the best model, which also partially coincides with this work. Similarly, in the case of the sorption isotherm of pine nuts (Nayab et al. 2023),

the mGAB model proved to be the best for the adsorption and desorption isotherms of the samples at individual temperatures; however, it failed to outperform other models in multi-temperature estimation. On the other hand, the mGAB model is still quite useful for determining the monolayer moisture content since its first parameter (A , originally indicated as M_0) corresponds to this value.

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Table 4. Parameters of the Guggenheim-Anderson-de Boer (mGAB), Halsey (mHAL), Henderson (mHEN), and Oswin (mOSW) models for dried tomato adsorption isotherms for the temperatures range of 15 to 35 °C determined in multi-temperature estimation

Parameters	Multi-temperature estimation for SIs in the range of 15 to 35 °C			
	mGAB	mHAL	mHEN	mOSW
A	0.160 ± 0.028	-13.933 ± 0.038	-0.036 ± 0.015	-1.328 ± 0.517
B	0.916 ± 0.024	0.038 ± 0.012	-367.710 ± 28.68	0.006 ± 0.002
C	$2.021 \times 10^{5*}$	2.041 ± 0.188	0.947 ± 0.116	2.255 ± 0.207

* The 95% confidence limit was not determined; SI – sorption isotherm

Table 5. Statistical goodness of fit indices of the Guggenheim-Anderson-de Boer (mGAB), Halsey (mHAL), Henderson (mHEN), and Oswin (mOSW) models for dried tomatoes adsorption isotherms for the temperature range of 15 to 35 °C determined in the multi-temperature estimation

Models	Multi-temperature estimation for SIs in the range of 15 to 35 °C			
	SSE	R^2	RMSE	AIC_c
mGAB	0.264	0.947	0.112	-98.09
mHAL	0.143	0.971	0.083	-112.80
mHEN	0.182	0.963	0.093	-107.04
mOSW	0.132	0.974	0.079	-114.81

SSE – sum of squares of errors; R^2 – coefficient of determination; RMSE – root mean square error; AIC_c – corrected Akaike information criterion values; SI – sorption isotherm

CONCLUSION

This study provides the results of SI modelling for traditional sun-dried tomatoes that have been dried at high altitudes, low pressure, relative humidity, and temperature of the surrounding atmosphere, which is typical for north-central Afghanistan. Modified sorption models can be applied to predict the $EMC-a_w$ relationships of dried tomatoes. From the evaluation of the SI models with incorporated temperature terms, it resulted that the mOswin, mHalsey, and Henderson models with the RMSE values of 0.079, 0.083, and 0.093, respectively, outperformed the mGAB model with RMSE = 0.112 in multitemperature SI evaluation. However, at individual temperatures, the mGAB model fitted the SI data of dried tomatoes the best, with the lowest SSE, RMSE, and AIC_c and the highest R^2 . The SIs of the dried tomato samples were classified as type III exponential SI.

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