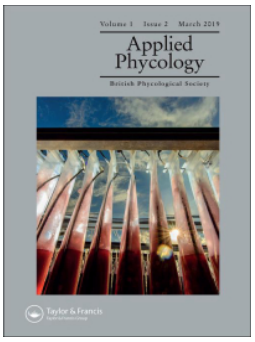

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The effect of natural convection air temperature on the drying kinetics and desorption isotherms of *Alaria esculenta* and *Palmaria palmata*

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ABSTRACT

The Northern Periphery and Arctic region faces unique economic and food security challenges that may be partly answered by commercial seaweed production. *Alaria esculenta* and *Palmaria palmata* are two seaweeds commonly found in the region and suitable for cultivation and processing for food and other commercial products. The drying kinetics for both species were obtained, and the Page and Weibull models best described the data. A drying air temperature increase from 40 to 70°C decreased drying time by 62.4% and 61.7% for *A. esculenta* and *P. palmata,* respectively. Desorption isotherms were obtained between 25 and 70°C and showed Brunauer Category III shapes, with water activity increasing with temperature for a fixed moisture content. Net heats of desorption were obtained, with drying to an equilibrium moisture content of 0.01 $kg_{water}\ kg_{d.b.}^{-1}$ requiring 18.1 and 3.94 kJ $mol^{-1}\ K^{-1}$ for *A. esculenta* and *P. palmata,* respectively.

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Convective blown air drying; desorption isotherms; drying kinetics; heat of sorption

Introduction

The Northern Periphery and Arctic (NPA) region, although geographically diverse, shares several challenges including low accessibility, low economic diversity and high potential impact of climate change (Northern Periphery and Arctic Programme, 2016). Population growth and climate change have the potential to exacerbate these issues and raise food security concerns. Additionally, growing awareness of the carbon footprint of foods raises calls for food produced locally, efficiently and sustainably. The NPA Programme aims to overcome such challenges by transnational project cooperation involving nine partner countries in the region.

Seaweed production may form part of the solution to these challenges. Seaweed cultivation has no land area demand, and a processing facility needs only a small land footprint; there is thus no competition with agriculture for viable grazing or arable land, which is scarce in many parts of the NPA region (Jacobson, 2016; Kintisch, 2016). Seaweed cultivation and processing offers an alternative or complementary livelihood to fishing using many of the same skills and not requiring major retraining.

SW-GROW is a multidisciplinary project funded by the NPA that aims to innovate and communicate research and best practice to bolster the seaweed industry in the region. The SW-GROW programme identified two seaweed species as of special interest for food cultivation: *Alaria esculenta* and *Palmaria palmata. A. esculenta* (winged kelp, dabberlocks or lair) is a brown seaweed found at low tide and in the sublittoral zone across the NPA region (Springer, Lütz, Lütz-Meindl, Wendt, & Bischof, 2017). Its regional limits are determined by the 16°C summer water temperature isotherm (Lüning, 1990); with warming seawater, its southern range is expected to contract (Mieszkowska et al., 2006); however, its abundance across the British Isles has not changed significantly between 1974 and 2010 (Yesson, Bush, Davies, Maggs, & Brodie, 2015). It is relatively tolerant of salinity variation (Fredersdorf, Müller, Becker, Wiencke, & Bischof, 2009). It has been used as a food for humans (Chapman, Stévant, & Larssen, 2015) and land (Seterlund, Hoie, Sannan, & Raastad, 1968) and sea (Mai, Mercer, & Donlon, 1994) dwelling herbivores, and contains viable quantities of sugars, proteins and minerals (Schiener, Black, Stanley, & Green, 2015). It can also be used for nutrient sequestration in aquaculture (Reid et al., 2013) and in the production of biocoal and biomethane by hydrothermal carbonization (Smith & Ross, 2016). To date various seeded rope methods have been investigated for commercial cultivation of *A. esculenta* (Kerrison et al., 2020).

P. palmata (dulse, dillisk, söl) is a red seaweed found in the littoral and sublittoral zones across the NPA region (Werner & Dring, 2011); growth rate deterioration begins between 14°C and 18°C summer

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temperature (Morgan & Simpson, 1981), and salinity. It is amongst the most commonly used seaweeds for human consumption – in the UK and Ireland, Norway, the Faroes, Iceland and New England – due partly to its attractive taste profile, and is relatively high in amino acid and lipid content (Mouritsen et al., 2013). Additionally, the long-chain lipids in *P. palmata* are more readily digestible to humans than those found in other seaweeds (Lopes et al., 2019; van Ginneken, Helsper, de Visser, van Keulen, & Brandenburg, 2011) and can be further enhanced by heat treatment (Maehre, Edvinsen, Eilertsen, & Elvevoll, 2016). Alongside food use, *P. palmata* supplements have may have health benefits including hypertriglyceridaemia reduction (Takase et al., 2020) and hypertension and Type II diabetes control (Harnedy & FitzGerald, 2013).

Both seaweed species can be eaten fresh, but, because fresh seaweed is highly perishable, dehydration is normally required for commercial distribution and in order to preserve the seaweed for future consumption. Drying is an energy-intensive step in seaweed processing, and the objectives are to maximise nutritional characteristics whilst preventing microbial growth, at the minimum energy cost. Microbial growth is promoted with increasing water activity but cannot occur where $a_w < 0.6$ (Fontana, 2020). Al-Muhtaseb, McMinn, and Magee (2002) provide a succinct review of water activity isotherm modelling. Historically, open air drying was common but convective air drying methods are common today; principally because they are relatively simple to design and effectively preserve nutrients in the dried seaweed (Badmus, Taggart, & Boyd, 2019; Uribe et al., 2019). Erbay and Icier (2010) and Onwude et al. (2016) present thorough reviews and classification of convective drying models. This paper provides the drying kinetics, desorption isotherms and isosteric desorption heats of *A. esculenta* and *P. palmata*. Additionally, it enables seaweed producers to design and calibrate convection drying systems in which convection is either natural or the air velocity is low. We expect to report the effects of air velocity, temperature and humidity in forced convective drying systems in a future paper.

Materials and methods

Sample preparation

Both seaweed species were wild harvested: *A. esculenta* from the coastline stretching from 58°15′02"N 6°08′23"W to 58°14′02"N 6°09′23"W, on the Eye Peninsula, Isle of Lewis, UK; and *P. palmata* from 57°49′07"N 6°49′52"W to 57°49′03"N 6°49′48"W at Stockinish, Isle of Harris, UK. Both species were harvested between April and July 2020 to obtain kinetics for the typical harvesting season. Fig 2 shows the seaweeds after harvesting and before sample preparation. Harvested seaweed was immediately placed in a sealed container filled with seawater and then refrigerated at 3 ± 1°C within 1 h of harvesting. Scoping experiments showed that *A. esculenta* deteriorated more rapidly than *P. palmata* and began to discolour and develop a sharp smell after ~80 h of refrigeration; therefore, any leftover seaweed was discarded after 72 h to ensure sample consistency.

When ready for use, samples of 2.5 ± 0.2 g by wet basis (w.b.) were prepared. Whilst *P. palmata* is a homogenous material, *A. esculenta* consists of a thick, dense stripe and a thin blade; therefore, care was taken to ensure that the planar stripe-to-blade ratio of each *A. esculenta* sample was approximately constant. The samples were rinsed in fresh water and then shaken for 5 s to remove surface water droplets. All drying and desorption tests were performed in triplicate and the mean taken of the results.

Drying

Drying was performed with a Kern DAB 100-3 moisture analyser that removed moisture from the samples by natural convection heating at a specified temperature. This was connected by RS-232 lead to a PC for automated data recording, as shown in Fig 1. The time-based drying curves were obtained with a temporal sampling resolution of 1 min and a mass accuracy of ± 0.5 mg. Each sample was considered dry ($MR = 0$) when the sample mass decreased by less than 10^{-5} g min^{-1} for five consecutive minutes.

The experimental moisture ratio is calculated as

$$MR = \frac{X - X_e}{X_0 - X_e}. \quad (1)$$

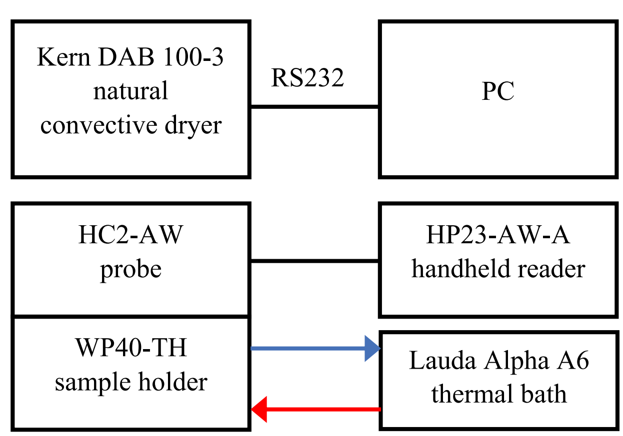

Figure 1. Block diagram to show the setup for the drying and desorption experiments.

Figure 2. Photographs with a 30 cm ruler of (a) *Alaria esculenta* and (b) *Palmaria palmata*, as harvested before trimming to sample weight.

Table 1. Drying models.

Model	Expression	Reference
Lewis–Newton	$MR = \exp(-kt)$ (7)	Lewis (1921)
Page	$MR = \exp(-kt^n)$ (8)	Page (1949)
Modified Page I	$MR = \exp\,(-kt)^n$ (9)	Overhults et al. (1973)
Weibull	$MR = \exp\left[-\left(\frac{t}{a}\right)^{\beta}\right]$ (10)	Weibull (1951)

This was compared with the empirical models shown in Table 1. Lewis (1921) model is often referred to in the literature as Newton's as it derives from Newton's law of cooling. Page (1949) added the empirical constant n, which was modified by Overhults, White, Hamilton, and Ross (1973).

The coefficient of determination is defined as

$$r^2 = 1 - \frac{SS_{res}}{SS_T}, \quad (2)$$

where sum-of-squares subscripts *res* and T denote residuals and total, respectively.

For each model in Table 1, the fitting constants were obtained by nonlinear regression using the Trust-Region Method in MATLAB.

Desorption

Water activity was measured with Rotronic water activity metering equipment: samples were placed inside a WP-40TH water-jacketed sample holder, on which sat an HC2-AW capacitive measurement probe connected to the HP23-AW-A handheld reader, returning a_w accurate to ± 0.01. The water-jacketed sample holder was fed by a thermostatically controlled Lauda Alpha A6 thermal bath.

To obtain the desorption isotherms, the samples were dried with the moisture analyser until the mass (± 0.025 g) for each data point was obtained, then transferred to the water activity meter, regulated to the desired temperature, and the water activity was read once stable for 5 min. Data points were obtained at relative humidities of [0.75, 0.5, 0.4, 0.3, 0.275, 0.25, 0.225 and 0.2] of the initial wet mass.

A comparison of the sorption models shown in Table 2 was made. The Guggenheim–Anderson– de Boer (GAB) model (Equation (11)) was derived independently by Anderson (1946), de Boer (1953), and Guggenheim (1966). It is a development of the Langmuir (1918) and Brunauer, Emmett, and Teller (1938) (BET) models and uses fitting parameters with physical meaning:

Table 2. Desorption models.

Model	Expression	Reference
Guggenheim–Anderson–de Boer (GAB)	$X_e = \frac{X_0 kCa_w}{(1-ka_w)(1-ka_w+kCa_w)}$ (11)	Anderson (1946), de Boer (1953), Guggenheim (1966)
Oswin	$X_e = A\left(\frac{a_w}{1-a_w}\right)^B$ (12)	Oswin (1946)
Halsey	$X_e = \left(\frac{-A}{\ln(a_w)}\right)^B$ (13)	Halsey (1948)
Caurie	$X_e = \exp\left(a_w \ln(A) - \frac{1}{4.5X_s}\right)$ (14)	Caurie (1970)

Table 3. Drying model parameters of *A. esculenta*.

Model	T (°C)	r^2	a	β
Weibull	40	.997	46.1	.863
	50	.998	33.2	.888
	60	.996	18.8	.855
	70	.991	12.2	.843
Model	T (°C)	r^2	$k \times 10^{-4}$	n
Lewis–Newton	40	.989	3.47	-
	50	.993	4.87	-
	60	.988	8.51	-
	70	.984	13.2	-
Page	40	.998	10.8	.863
	50	.998	11.8	.888
	60	.996	24.7	.855
	70	.992	38.6	.843
Modified Page	40	.989	6.40	.542
	50	.993	10.3	.471
	60	.988	11.3	.751
	70	.984	18.9	.700

$$C = c_0 \exp\left(\frac{H_m - H_n}{RT}\right), \tag{3}$$

$$k = k_0 \exp\left(\frac{H_l - H_n}{RT}\right), \tag{4}$$

where c_0 and k_0 are entropic fitting parameters and subscripts *m, n*, and 1 denote monolayer, multilayer and liquid, respectively. This was compared with the empirical models of Oswin (1946), Halsey (1948) and Caurie (1970). When evaluating these models, data points where $a_w > 0.95$ were excluded as the models are not established for water activity values approaching unity.

The isosteric heat of sorption is:

$$Q_{st} = q_{st} + H_{fg}, \tag{5}$$

where H_{fg} is the latent heat of vaporization of water at the sample temperature. The net isosteric heat of sorption is given by the Clausius–Clapeyron equation:

$$q_{st} = -R\frac{\partial \ln a_w}{\partial 1/T}\bigg|_{X_e}, \tag{6}$$

evaluated at fixed moisture content.

Results

Drying kinetics

Fig 3 shows the logarithmic drying curves for the seaweed species studied. For both species, the layer half-thickness was 0.75 mm. Across all drying temperatures, *A. esculenta* took longer to dry than *P. palmata* and the temperature effect was of the same order of magnitude: increasing air temperature from 40 to 70°C decreased total drying time by 62.4% and 61.7% for *A. esculenta* and *P. palmata*, respectively. Tables 3 and 4 show the drying model parameters for *A. esculenta* and *P. palmata*, respectively. All the curves are acceptably modelled by exponential functions; of the semi-empirical models, Page returned the highest mean r^2 value. The Weibull model also returned excellent r^2 values for both seaweeds. The β parameter was consistently lower for *A. esculenta* than *P. palmata*, indicating a drying curve that has a steeper initial profile but also levels off sooner (Bantle, Kolsaker, & Eikevik, 2011).

Desorption isotherms

Figs 4 and 5 provide the desorption isotherms for *A. esculenta* and *P. palmata*, respectively; water activity was plotted on the *x*-axis according to convention, although it is the dependent variable in these tests. The "dry" datapoints corresponding to $X_e = 0\ kg_{water}\ kg^{-1}$ d.b.

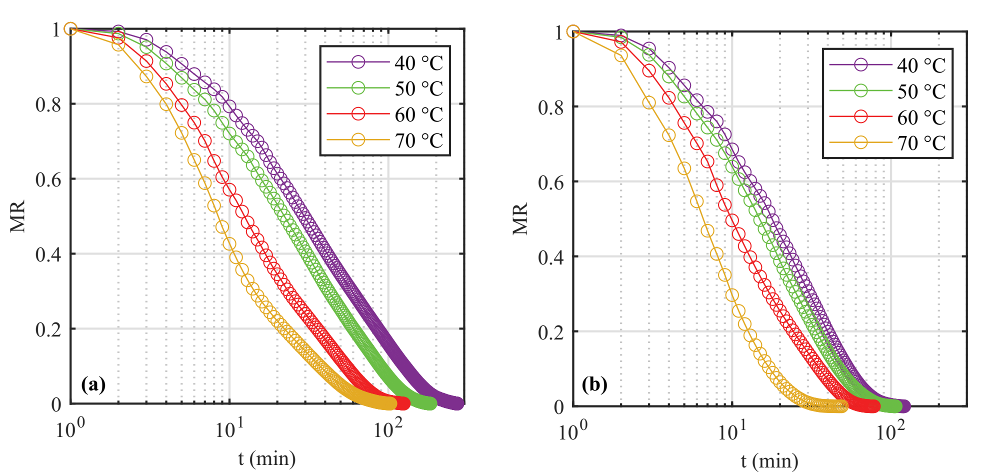

Figure 3. Effect of air temperature on drying curves of (a) *Alaria esculenta* and (b) *Palmaria palmata*.

Table 4. Drying model parameters of *Palmaria palmata*.

Model	*T* (°C)	r^2	*α*	*β*
Weibull	40	.999	24.1	1.06
	50	.999	20.2	1.03
	60	.997	13.8	.973
	70	1	7.76	1.16
Model	***T* (°C)**	r^2	$k \times 10^{-4}$	*n*
Lewis-Newton	40	.998	7.02	-
	50	.999	8.32	-
	60	.997	12.0	-
	70	.995	22.1	-
Page	40	.999	4.42	1.06
	50	.999	6.70	1.03
	60	.995	7.50	1.07
	70	1.00	8.09	1.16
Modified Page	40	.998	12.6	.559
	50	.999	16.6	.501
	60	.997	17.4	.687

Table 5. Desorption model parameters of *Alaria esculenta*.

Model	*T* (°C)	r^2	Fitted constants
GAB (X_0 in units $kg_{water}\ kg_{d.b.}^{-1}$)	25	.909	$X_0 = 3.54$, $k_b = .946$, $C = 0.01316$
	40	.978	$X_0 = 3.55$, $k_b = .963$, $C = 0{,}004594$
	50	.897	$X_0 = 1.62$, $k_b = .865$, $C = 0.0071$
	60	.962	$X_0 = 1.02$, $k_b = 1$, $C = .00458$
	70	.790	$X_0 = 2.19$, $k_b = 1$, $C = 0.00268$
Oswin	25	.911	$A = 0.0698$, $B = 1.51$
	40	.975	$A = 0.0275$, $B = 1.52$
	50	.884	$A = 0.0165$, $B = 1.11$
	60	.976	$A = 4.73 \times 10^{-3}$, $B = 2.14$
	70	.862	$A = 3.23 \times 10^{-3}$, $B = 2.55$
Halsey	25	.904	$A = 0.161$, $B = 1.78$
	40	.971	$A = 0.0900$, $B = 1.67$
	50	.877	$A = 0.0261$, $B = 1.20$
	60	.977	$A = 0.0794$, $B = 2.39$
	70	.859	$A = 0.103$, $B = 2.85$
Caurie	25	.923	$A = 2188$, $X_s = 0.0343$
	40	.995	$A = 5.68 \times 10^4$, $X_s = 0.0220$
	50	.931	$A = 4457$, $X_s = 0.0240$
	60	.965	$A = 7.93 \times 10^5$, $X_s = 0.0167$
	70	.868	$A = 2.97 \times 10^6$, $X_s = 0.0158$

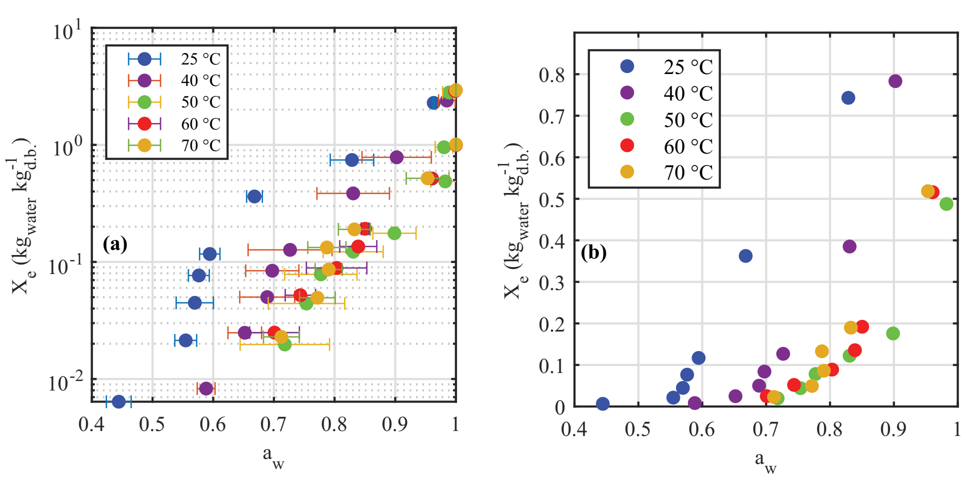

Figure 4. Desorption isotherms of *Alaria esculenta* showing (a) full isotherms in log scale and (b) detail at low moisture content.

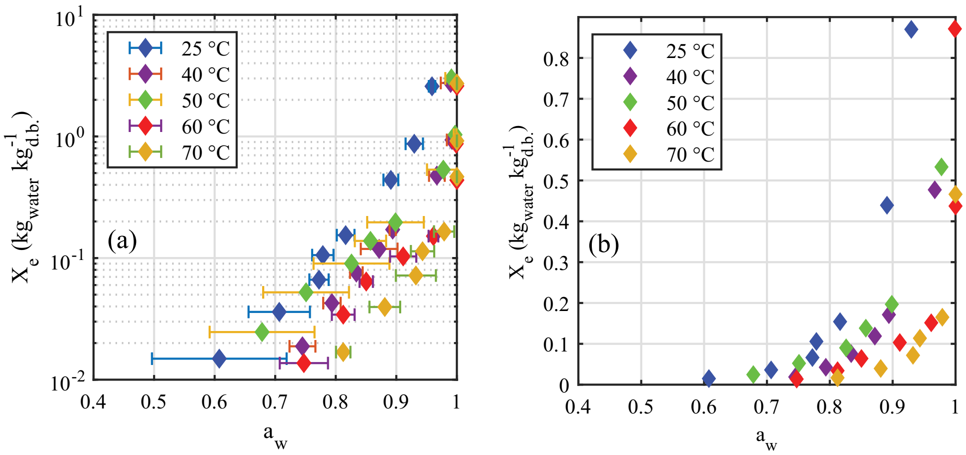

Figure 5. Desorption isotherms of *Palmaria palmata* showing (a) full isotherms in log scale and (b) detail at low moisture content.

Table 6. Desorption model parameters of *Palmaria palmata*.

Model	T (°C)	r^2	Fitted constants
GAB (X_0 in units kg$_{water}$ kg$_{d.b.}^{-1}$)	25	.996	X_0 = .458, k_b = .988, C = 0.0165
	40	.990	X_0 = .116, k_b = .992, C = 0.0252
	50	.989	X_0= .171, k_b = .935, C = 0.0431
	60	.946	X_o = 0.0127, k_b = 1, C = 0.281
	70	.963	X_0 = .00990, k_b = 1, C = 00932
Oswin	25	.995	A = 0.0138, B = 1.61
	40	.993	$A = 4.04 \times 10^{-3}$, B = 1.76
	50	.984	A = 0.0135, B = 1.24
	60	.942	$A = 6.31 \times 10^{-3}$, B = 1.21
	70	.967	$A = 2.04 \times 10^{-3}$, B = 1.41
Halsey	25	.994	A = 0.0678, B = 1.72
	40	.991	A = 0.0447, B = 1.91
	50	.980	A = 0.0328, B = 1.35
	60	.937	A = 0.0166, B = 1.30
	70	.968	A = 0.0130, B = 1.48
Caurie	25	.996	$A = 3.14 \times 10^{6}$, X_s = 0.0158
	40	.997	$A = 1.14 \times 10^{6}$, X_s = 0.0156
	50	.994	$A = 1.29 \times 10^{4}$, X_s = 0.0219
	60	.975	$A = 2.99 \times 10^{4}$, X_s = 0.0191
	70	.929	$A = 1.66 \times 10^{6}$, X_s = 0.0141

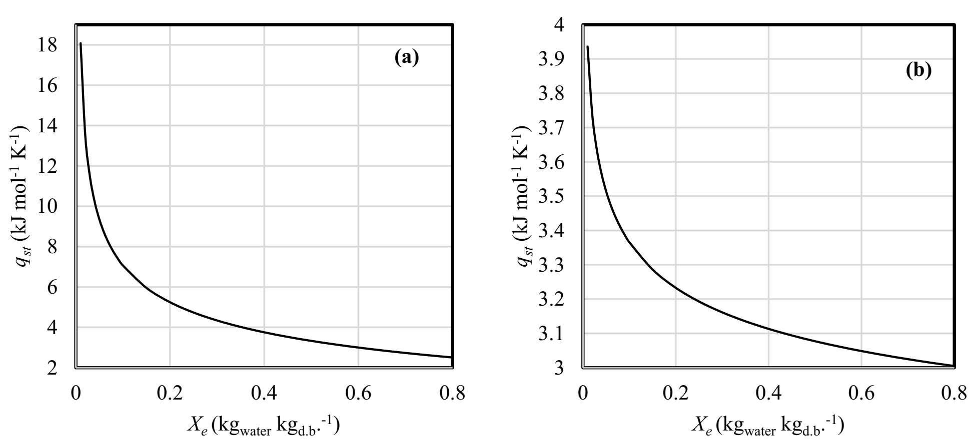

Figure 6. Net isosteric heat of desorption for (a) *Alaria esculenta* and (b) *Palmaria palmata*.

were not plotted; the water activities jumped notably from their previous point (as graphed) to $a_w < 0.5$ when moisture content moved from the previous non-zero X_e to zero. From Figs 4(b) and 5(b), both species exhibit Category III isotherms according to the Brunauer classification (Brunauer, Deming, Deming, & Teller, 1940). Across the moisture contents tested, increasing temperature increased the measured water activity. Tables 5 and 6 show that all four sorption models evaluated fitted the data well, although the Oswin and Halsey models performed marginally poorer than the Caurie model. Previous seaweed desorption studies have not observed a significant performance variation between models tested (Arufe, Torres, Chenlo, & Moreira, 2018; Moreira, Chenlo, Sineiro, Sánchez, & Arufe, 2016).

To obtain the isosteric heat of sorption, the Caurie model was used to calculate water activity for a given moisture content, and then substituted into Equation (6) to return the results shown in Fig 6 for *A. esculenta* and *P. palmata,* respectively. This shows that drying to an equilibrium moisture content of 0.01 $kg_{water}\ kg_{d.b.}^{-1}$, *A. esculenta* requires 4.59× the energy of *P. palmata*.

Nomenclature

A	Sorption fitting constant
a_w	Water activity
B	Sorption fitting constant
C	GAB fitting constant
H	Molar sorption enthalpy
$H\{fg\}$	Latent heat of vaporization
k	Constant
MR	Moisture ratio
n	Empirical constant
Q_{st}	Isosteric heat of sorption
q_{st}	Net isosteric heat of sorption
R	Gas constant = 8.314
r^2	Correlation coefficient
SS	Sum-of-squares
T	Temperature
t	Time
X	Instantaneous moisture content
X_0	Monolayer moisture content
X_e	Equilibrium moisture content
X_s	Security moisture content
α	Weibull coefficient
β	Weibull coefficient

Disclosure statement

No potential conflict of interest was reported by the author(s).

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