EQUILIBRIUM MOISTURE CONTENT OF HAZELNUT HUSKS, SHELLS, AND KERNELS

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ABSTRACT. *Hybrid hazelnuts that are predominately a cross between the American hazelnut (*Corylus americana*) and the European hazelnut (*Corylus avellana*) are being grown and evaluated as part of an effort to develop a thriving hazelnut industry for the Upper Midwest of the U.S. Along with this plant development effort, researchers are investigating and assessing various harvesting and processing methods and equipment in an effort to create a robust and food-safe production industry. One harvesting alternative is to pick hazelnut clusters off plants before the nuts fully ripen and fall to the ground, an approach that requires greater attention to drying. Whether entire clusters are dried or the nuts are separated from the husks prior to drying is a decision that will be influenced by the drying requirements and potential uses for these hazelnut fractions. To this end, a study was undertaken to establish desorption isotherms for the husks, shells, and kernels of hybrid hazelnuts grown in the Upper Midwest. Clusters were hand-picked from shrubs in Wisconsin and immediately placed in 18 different controlled environments (six different relative humidity levels at three different temperatures). Actual moisture conditioning took place over saturated salt solutions in specially fabricated biomaterial moisture conditioning units. After a six-week period during which the clusters reached equilibrium with their environment via desorption, they were separated into husk, shell, and kernel fractions and returned to their respective conditioning units. After another six weeks in the conditioning units, the moisture content (MC) of each fraction was determined by oven-drying at 103°C for 48 h. Under equilibrium conditions, the kernel MC was found to be only 37% of that for shells, whereas the equilibrium moisture content (EMC) values for husks were on average 14% greater than those for shells. On a dry basis, the average cluster mass was 32.9% husk, 43.9% shell, and 23.2% kernel. Likewise, on a dry basis, the average whole nut mass was 65.5% shell and 34.5% kernel. The desorption data were fit to the Modified Henderson, Modified Chung-Pfost, Modified Halsey, Modified Oswin, and Modified GAB equations. Overall, the best fit to the experimental data was provided by the Modified Chung-Pfost equation with parameters determined using equilibrium relative humidity (ERH) as the dependent variable in regression analyses. For ERH values above 0.70, the temperature-modified form of the GAB equation is recommended for predicting desorption EMC values for hazelnut fractions.*

Keywords. *Desorption, Equilibrium moisture content, Equilibrium relative humidity, Hazelnuts, Kernels, Nuts, Shells, Water activity.*

The Upper Midwest Hazelnut Development Initiative (UMHDI) is a group of hazelnut growers and researchers primarily located in Wisconsin and Minnesota with a goal of developing a hazelnut industry in the U.S. Upper Midwest. Fischbach (2017) listed the four primary objectives of the UMHDI as developing: (1) hazelnut germplasm capable of supporting an economically viable Upper Midwest industry, (2) propagation procedures capable of producing low-cost clonal material, (3) harvesting and processing equipment tailored to the selected germplasm, and (4) a growing and processing network supported by a robust outreach education program.

Development of hazelnut germplasm for the Upper Midwest has primarily focused on crosses between the European hazelnut (*Corylus avellana*) and the American hazelnut (*Corylus americana*). *C. avellana* grows naturally as a large shrub (3 to 8 m tall) but is commonly pruned to grow as a tree when cultivated for nut production. Cultivated genotypes of *C. avellana* are very productive and produce measurably larger nuts than *C. americana*. However, because of its susceptibility to Eastern Filbert Blight (EFB), *C. avellana* has not been cultivated east of the Rocky Mountains. *C. americana* grows as a medium to large shrub (2.5 to 5 m tall) and is native to portions of the U.S. east of the Great Plains. Researchers working with crosses of *C. avellana* and *C. americana* are intent on identifying genotypes with the productivity of *C. avellana* and the EFB resistance and cold-hardiness of *C. americana* (Braun et al., 2019).

Hazelnuts grow in clusters, with each nut in the cluster surrounded by an involucre (fig. 1). A more commonly used (but less technical) term for an involucre is a husk. As nuts reach maturity, nutrient flow to the clusters is curtailed, the involucres (husks) lose moisture content and change from light green to light brown, and the shell surface changes from white to brown. Left to fully ripen, the nuts (and in some cases entire

Submitted for peer review in December 2018 as manuscript number PRS 13257; approved for publication as a Research Article by the Processing Systems Community of ASABE in June 2019.

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Vol. 62(5): 1075-1086 ISSN 2151-0032 https://doi.org/10.13031/trans.13257

Figure 1. Variation in hazelnut cluster maturity.

clusters with nuts still attached) will be released from the plant and fall to the orchard floor. The nuts are then collected using equipment compatible with the slope, roughness, and amount of vegetation on the orchard floor.

Unlike farmers elsewhere, the vast majority of farmers growing hazelnuts in the Upper Midwest do not harvest nuts off the orchard floor; instead, they pick hazelnut clusters from the plants before the nuts are fully ripened. These early-harvested clusters are herein referred to as "green clusters". Harvesting green clusters is made more possible by the smaller size of *C. americana* and its hybrids, and it reduces nut loss to animal predation, eliminates nut contamination associated with orchard floor contact, and eliminates mold growth that can occur under prolonged damp conditions on the orchard floor.

A negative aspect of harvesting green clusters is the additional equipment, labor, and energy required to dry the clusters to a point they would reach naturally if left on the plant. Uniform cluster drying is made more difficult by the tight manner in which clusters pack, a situation that largely restricts drying to thin layers. In addition to investigating green cluster harvesting using over-the-row mechanical shakers, on-going research at the University of Wisconsin-Madison is focused on green cluster husking, which is the immediate separation of husks from nuts. The advantages of green cluster husking are that it eliminates the cost associated with removing moisture from the husks, and the subsequent drying of nuts without husks is not restricted to thin layers.

Regardless of whether entire clusters or just nuts are dried, the drying must be performed at a rate and under conditions that stave off mold growth but do not induce drying stresses that crack the shells, thereby exposing the kernels to possible contamination. Determining such ideal drying rates and conditions for a hydroscopic material requires knowledge of the moisture content (MC) at which the material will be in thermodynamic equilibrium with its surrounding environment. This MC level, referred to as the material's equilibrium moisture content (EMC), is a function of the material properties, direction of material MC change (adsorption or desorption), and water vapor pressure of the surrounding gas. When the surrounding gas is air, the water vapor pressure is commonly expressed as a function of the air temperature and relative humidity (RH). For a given material MC and temperature, the RH at thermodynamic equilibrium (i.e., at the point of net zero water exchange between the material and surrounding air) is defined as the equilibrium relative humidity (ERH). A plot of EMC versus ERH for a given temperature is referred to as a sorption isotherm, or more specifically, a desorption isotherm if thermodynamic equilibrium is reached by decreasing the material MC and an adsorption isotherm if equilibrium is reached by increasing the material MC.

A review of scholarly publications reveals a vast amount of sorption isotherm research involving agricultural products and foods. Much of this research has been conducted since Wolf et al. (1985) compiled, indexed, and published a bibliography of over 2200 different documents on the subject. A good source for sorption isotherm data is ASABE Standard D245.6 (ASABE, 2012a). The data in ASABE Standard D245.6 are compiled in separate tables for (1) starchy materials, (2) fibrous materials and selected feedstuffs, (3) materials high in oil and protein, and (4) agricultural by-products. ASABE Standard D245.6 does not contain sorption isotherm data for hazelnut constituents or products.

From a material perspective, a hazelnut cluster consists of three distinct substances: husks, shells, and kernels. Because of their compositional differences, each of these substances has its own unique EMC-ERH relationship, meaning that each substance is at a different MC when the cluster is in thermodynamic equilibrium with the surrounding environment. Research shows that sorption isotherms for a material comprised of distinct fractions can be calculated from the sorption isotherms for the constituent fractions. More specifically, Ondier et al. (2012) showed that the EMC of rice could be calculated from the weighted averages of the EMC values of the constituent kernel fractions. Likewise, using EMC data published by Mazza and Jayas (1991), it was found that, for ERH values less than 70% and temperatures of 40°C and lower, there was no significant difference between the experimentally determined EMC for sunflower seeds and the EMC calculated using weighted averages of the EMC values for the kernels and hulls comprising the seeds.

A primary driving force for much of the sorption isotherm

research is the safe storage of food for human and animal consumption. Safe storage is largely achieved with an environment that does not support microbial growth. This is effectively accomplished with storage in an environment maintained at an RH of less than 65% or by ensuring that, prior to sealing into packaging, the product is at a moisture content below the EMC associated with an ERH of 65%. This ERH is about the driest environment in which xerophilic molds can survive (Fennema, 1996), as shown in figure 2.

Hazelnut kernels, unlike hazelnut husks and shells, have a high oil content. Using a Soxhlet extractor, researchers at the University of Nebraska (Xu et al., 2007) extracted oil from 25 different hybrid hazelnuts (i.e., interspecific crosses of *C. americana*, *C. avellana*, and *C. cornuta*) grown in southeastern Nebraska and found that the oil content ranged from 51.4% to 75.1% of dry kernel weight. Because of the hydrophobic nature of such oils, the EMC for hazelnut kernels at a given temperature and RH can be expected to be measurably less than the EMC values for hazelnut husks and shells. Researchers working with sunflower seeds (Mazza and Jayas, 1991; Maciel et al., 2015) all reported a decrease in EMC with increasing oil content; however, Mazza and Jayas (1991) also noted that when calculated on an oil-free basis, seeds of varying oil content yielded nearly the same EMC-ERH relationship. Maciel et al. (2018) developed a plot of safe storage moisture contents for sunflower seeds as a function of oil content and temperature.

Several theoretical, semi-empirical, and empirical models have been developed to describe EMC-ERH relationships (Al-Muhtaseb et al., 2002; Andrade et al., 2011; Limousin et al., 2007); collectively, these models are generally referred to as sorption isotherm equations. ASABE Standard D245.6 (ASABE, 2012a) uses five such equations to define EMC-ERH relationships for plant-based agricultural products: the Modified Henderson equation (Thompson et al., 1968), the Modified Chung-Pfost equation (Pfost et al., 1976), the Modified Halsey equation (Iglesias and Chirife, 1976), the Modified Oswin equation (Chen, 1988), and the Guggenheim-Anderson-deBoer (GAB) equation (Van der Berg and Bruin, 1981).

Because of the current lack of sorption isotherm data for hazelnuts, the importance of such data for drying of green clusters and nuts, and for the safe storage of such nuts, a study was undertaken to establish moisture desorption isotherms for hazelnut husks, shells, and kernels and to model these desorption isotherms with the equations promoted in ASABE Standard D245.6.

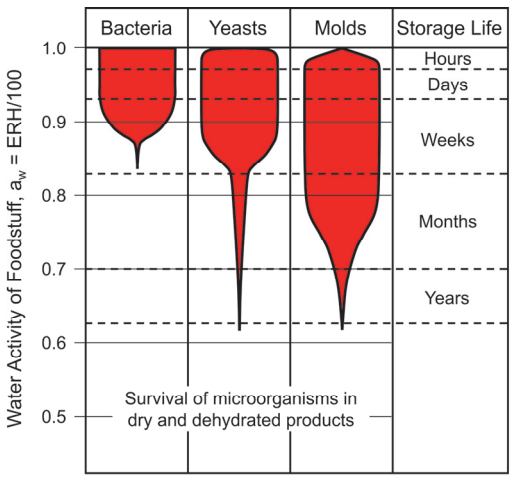

Figure 2. Diagram of microbial activity in foodstuffs and associated foodstuff storage life as a function water activity. Adopted from Richard-Molard et al. (1985).

Materials and Methods

Hazelnut clusters (fig. 1) were hand-picked on 1 September 2016 from hazelnut shrubs located on a farm near Stoughton, Wisconsin. This particular planting was established in 2011 using full-sibling F1 seedlings from a controlled-cross block managed by Forest Agriculture Enterprises, Viola, Wisconsin (Fischbach and Tibbals, 2016). The parents for the controlled cross were selected by Forest Agricultural Enterprises from their own mature planting of hybrid hazelnuts.

Harvested clusters were immediately transported from the field to the University of Wisconsin-Madison Agricultural Engineering Lab and mixed to more thoroughly blend clusters from different shrubs. All clusters used in this study had green or mostly green husks, which (based on hazelnut harvesting research at UW-Madison) typically indicates a cluster moisture content above 50% wet basis. Given their green state, all clusters were expected to reach equilibrium in all target environments via desorption.

A static gravimetric method employing saturated salt solutions was then used to obtain EMC versus ERH relationships at three different temperatures. This was accomplished with the use of 18 biomaterial moisture conditioning units. As shown in figure 3, each of these units consists of two HDPE FDA-compliant 5 gal (19 L) pails. Biomaterials (in this case, hazelnut clusters) were placed in the top pail, and a saturated salt solution was placed in the bottom pail. These two pails are herein referred to as the biomaterial pail and the salt solution pail, respectively. A 3.6 W fan constantly circulated air down over the surface of the saturated salt solution and back up through the biomaterial. For design details and an assessment of the biomaterial moisture conditioning units, see Bohnhoff (2017).

Six different salts were used in this study: magnesium chloride (MgCl), sodium acetate ($C_2H_3NaO_2$), calcium chloride ($CaCl_2$), potassium carbonate (K_2CO_3), sodium chloride (NaCl), and potassium chloride (KCl). Between 0.5 to 1.0 kg of tap water was saturated with salt and placed in the salt solution pails. Each pail was then labeled with the type of salt it contained (e.g., a $CaCl_2$ pail is shown in fig. 3). Three salt solution pails were prepared using each salt, one for each of the three temperatures investigated.

Approximately 8 L of hazelnut clusters were placed in each biomaterial pail. A cover was then snapped onto the pail, the biomaterial pail was weighed with a Sartorius LC34 laboratory scale, and the pail was then set into a salt solution pail. Six units (one for each of the six salt types) were placed in each of three temperature-controlled rooms and connected to a 12 VDC power supply. Precision glass thermometers were suspended through the cover of two biomaterial pails in each room. These thermometers were checked on a regular basis to ensure that the desired temperature was maintained within

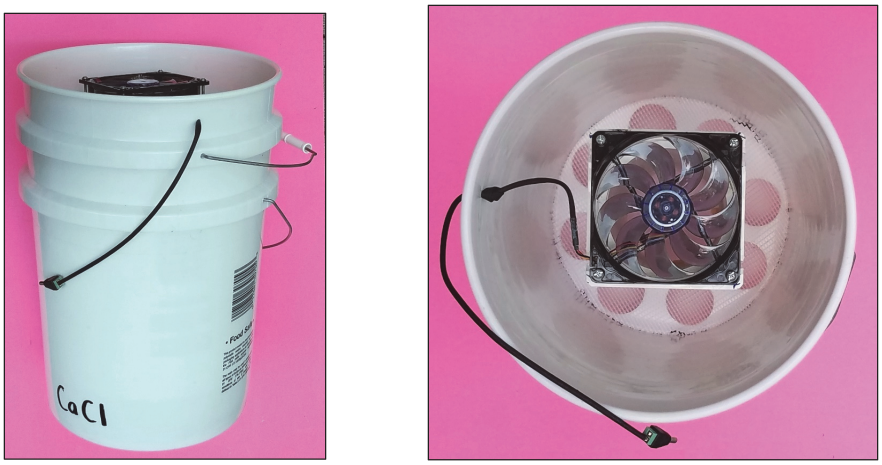

Figure 3. Side and top views of a biomaterial moisture conditioning unit (shown without snap-on cover).

each room.

Progress toward equilibrium was checked every other day for the first week. This simply involved disconnecting the biomaterial pail from the power supply, lifting the biomaterial pail out of its salt solution pail, and weighing both pails. Weighing of the pails was done in the room, with the biomaterial pail cover in place, and within a manner of seconds to avoid significant disturbance of the air within the pails. In almost all cases, the weight loss from the biomaterial pail was less than 1% different from the weight gain of the salt solution pail, an indication that the entire system was relatively leakproof. During each weighing, the degree of saturation and the level of the salt solution were checked. In several instances, a portion of the solution had to be removed from the salt solution pail, and the pail reweighed, because an increase in the solution volume (due to hazelnut cluster dehydration) threatened to block air circulation within the system.

Because the change in biomaterial pail weights between weighing intervals dropped off significantly after the first few days, the weighing interval was increased to a week. Between the fifth and sixth weeks, there was no change in weight for most pails, so the clusters were removed from each biomaterial pail and separated in husks, shells, and kernels. Each of these fractions was placed in a drying tin, and the tins were placed back inside the biomaterial pail from which the fraction originated. These tins were left uncovered in the sealed biomaterial pails for another six weeks, although no weight change was detected after the second week. This extra six-week period was allotted to compensate for moisture content changes that may have occurred when the clusters were broken into fractions, and to allow extra time for moisture located deep inside the kernels to diffuse to the kernel surface. After this second six-week period, the samples were removed from the biomaterial pails, and their moisture contents were determined.

For consistency, the moisture contents of all hazelnut constituents (husks, shells, and kernels) were determined in the same manner: by oven-drying at 103°C for 48 h. This is the same drying temperature and duration used by Palipane and Driscoll (1992) for macadamia nuts. The actual drying duration could have been reduced for husks and shells. Husks are similar in composition to forage crops, and ASABE Standard S358.3 (ASABE, 2017) specifies that forages be oven-dried at 103°C for 24 h. ASTM Standard D4442-16 (ASTM, 2016) specifies that wood and wood-based materials be oven-dried at 103°C with a duration that is based on the sample size and scale precision. From experience with small wood samples like hazelnut shells, this duration seldom exceeds 48 h. The 48 h duration used in this study was less that the 72 h at 103°C prescribed in ASABE Standard S352.2 (ASABE, 2012b) for edible beans, corn, and soybeans. However, compared to the outer cuticle on most bean seeds and the pericarp of corn kernels, the pellicle on the surface of hazelnut kernels is relatively vapor-permeable, thereby justifying a somewhat reduced drying time. Additionally, from previous lab work involving hazelnuts, moisture content specimen weights generally plateaued between 1 and 3 days after placement in the oven. With respect to kernels, it was felt that the drying time should not be excessive in order to minimize volatilization of non-aqueous substances (Bohnhoff, 2017).

A week prior to removing samples from a pail for moisture content determination, two Omega OM-141 USB mini data loggers were placed alongside the samples to record the temperature and RH within the pail. The data from these units were used in all subsequent analyses. Consequently, published ERH values for saturated salt solutions, as published in ASTM Standard E 104-2 (ASTM, 2017) and by Greenspan (1977), were not required. This procedure did not require the use of distilled water or high-purity inorganic salts. Instead, tap water and inexpensive salts, sold locally and in large quantities for water softening and road/highway deicing, were used.

Saturated Salt Solution ERH

The temperature and RH data collected with the Omega OM-141 dataloggers (after equilibrium had been attained within each biomaterial pail) were plotted for each of the different saturated salt solutions. Over the roughly 40°C range in temperature used in this study, the ERH for five of the saturated salt solutions was found to be a linear function of temperature. Regression of these data produced the following equations, which were subsequently used to calculate the ERH values:

$$\text{MgCl ERH} = 0.341 - 0.000571(T, °C) \quad (1)$$

$$K_2CO_3\ ERH = 0.454 - 0.000439(T, °C) \quad (2)$$

$$NaCl\ ERH = 0.761 - 0.000223(T, °C) \quad (3)$$

$$CaCl_2\ ERH = 0.774 - 0.001086(T, °C) \quad (4)$$

$$KCl\ ERH = 0.894 - 0.002137(T, °C) \quad (5)$$

The preceding equations for MgCl, NaCl, and KCl produced numbers very near those compiled by Greenspan (1977) and then adopted in ASTM Standard E104-2. The ERH values for K_2CO_3 aligned much better with those published by Labuza et al. (1985) than with those from Greenspan (1977). The linear equation for $CaCl_2$ should likely be dismissed. Although it yielded good values for the three temperatures used in this study, Young (1967) reported transitions occurring from $CaCl_2$ hexahydrate to $CaCl_2$ tetrahydrate at 29.4°C and from $CaCl_2$ tetrathydrate to $CaCl_2$ dihydrate at 45.5°C. In general, the relationship between temperature and RH tends to be very nonlinear at such transition points. This is likely the reason why $CaCl_2$ is seldom used to make saturated salt solutions for moisture sorption studies. That said, if RH is measured at equilibrium (as was done in this study), then the transitions of hydrates should not be an issue. In the end, when establishing sorption isotherms, it makes no difference how water vapor pressure is generated in an enclosure or what the RH level is; it just needs to be accurately recorded.

Sodium acetate ($C_2H_3NaO_2$, also CH_3COONa, NaOAc, and $MeCO_2Na$) was another salt used in this study that is seldom used by researchers to establish moisture sorption isotherms. The reason for this is unclear. From Young (1967), the salt is shown to exist in solution as sodium acetate trihydrate and have a linear ERH versus temperature relationship between 20°C and 40°C that can be approximated as:

$$C_2H_3NaO_2\ ERH = 0.850 - 0.0048(T, °C) \quad (6)$$

This equation yields ERH values of 0.846, 0.743, and 0.662 for the temperatures of 1.7°C, 23.3°C, and 40.0°C used in this study. These calculated values compare with the measured values of 0.795, 0.756, and 0.819. The value from equation 6 and the measured value at 23.3°C differed by 1.3%. This difference is greater at 1.7°C, which can be partially explained by the fact that equation 6 should only be used down to 20°C. The large difference at 40°C is believed to be due to the sodium acetate solution not being fully saturated when equilibrium was reached. A similar situation occurred with the magnesium chloride solution at 40°C. At equilibrium, the RH in the MgCl biomaterial pail was measured as 0.500. Equation 1 (which is based on measurements made over MgCl solutions known to be saturated at other temperatures) predicts an RH value of 0.318 at 40°C, which differs from the Greenspan (1977) value by only 0.2%. A visual check of the MgCl salt solution at 40°C after the completion of this study confirmed that the solution was not saturated. That the MgCl solution became unsaturated was not surprising, given the amount of MgCl required to form a saturated solution of the salt, and the amount of water that a saturated solution of MgCl is able to extract from a high-moisture product.

The situation with the unsaturated solutions of MgCl and $C_2H_3NaO_2$ at 40°C indicates the need to constantly check solutions when basing ERH values on published literature values instead of real-time measurements. Conversely, it also drives home the point that, when the RH is constantly monitored, non-saturated salt solutions can be used to establish moisture sorption isotherms.

RESULTS AND DISCUSSION

Husk, shell, and kernel EMC values for the 18 different combinations of temperature and RH are compiled in table 1. The EMC values for clusters in table 1 were calculated from the EMC values for husks, shells, and kernels using the mass fractions given in table 2. Likewise, the EMC values for whole nuts in table 1 were obtained by using the EMC values for kernels and shells along with their relative mass ratios from table 2.

DESORPTION ISOTHERMS

Experimental EMC, ERH, and temperature data were fit to

Table 1. Equilibrium moisture content values for hazelnut husks, shells, kernels, clusters, and whole nuts.

		Equilibrium Relative Humidity	Equilibrium Moisture Content (EMC, percent dry basis)				
Temperature	Salt	(ERH, decimal)	Husk	Shell	Kernel	Cluster[a]	Whole Nut[b]
1.7°C (35°F)	MgCl	0.340	12.7	11.6	4.8	10.4	9.2
	K_2CO_3	0.453	15.3	14.2	4.8	12.3	10.8
	NaCl	0.761	22.9	21.9	8.0	19.4	17.5
	$CaCl_2$	0.772	23.7	22.6	7.6	19.5	17.4
	$C_2H_3NaO_2$	0.795	26.5	23.5	8.1	20.6	17.9
	KCl	0.891	42.0	27.5	10.1	28.3	21.1
23.3°C (74°F)	MgCl	0.328	9.7	8.2	3.3	7.7	6.7
	K_2CO_3	0.444	11.1	10.8	4.2	9.1	8.2
	$CaCl_2$	0.735	19.5	18.4	5.4	16.1	14.0
	NaCl	0.749	19.7	18.9	6.6	16.4	14.9
	$C_2H_3NaO_2$	0.756	19.8	19.3	6.7	16.3	14.9
	KCl[c]	0.844	28.2	22.2	8.8	21.5	18.0
40.0°C (104°F)	MgCl	0.500	10.0	9.1	3.6	8.1	7.2
	K_2CO_3	0.436	8.7	8.0	3.3	6.9	6.3
	$CaCl_2$	0.731	18.1	15.3	5.8	14.4	12.2
	NaCl	0.752	18.5	15.6	5.4	14.2	12.3
	$C_2H_3NaO_2$	0.819	19.9	16.9	5.8	15.3	13.1
	KCl[d]	0.809	23.4	17.8	6.8	17.0	13.7

[a] Cluster = husk + shell + kernel.
[b] Whole nut = shell + kernel.
[c] Kernels were moldy.
[d] Husks and kernels were moldy.

Table 2. Cluster dry mass composition.

Temperature	Salt	Equilibrium Relative Humidity (ERH, decimal)	Percent of Oven-Dry Cluster Mass			
			Husk	Kernel	Shell	Whole Nut[a]
1.7°C (35°F)	MgCl	0.340	35.2	23.1	41.7	64.8
	K_2CO_3	0.453	34.6	23.9	41.5	65.4
	NaCl	0.761	35.2	20.9	43.9	64.8
	$CaCl_2$	0.772	32.7	23.3	44.0	67.3
	$C_2H_3NaO_2$	0.795	31.3	24.9	43.8	68.7
	KCl	0.891	34.3	24.2	41.6	65.7
23.3°C (74°F)	MgCl	0.328	33.6	20.8	45.6	66.4
	K_2CO_3	0.444	30.7	26.5	42.8	69.3
	$CaCl_2$	0.735	32.0	22.1	45.9	68.0
	NaCl	0.749	29.0	24.5	46.4	71.0
	$C_2H_3NaO_2$	0.756	39.1	20.7	40.2	60.9
	KCl[b]	0.844	34.4	20.7	44.9	65.6
40.0°C (104°F)	MgCl	0.500	32.7	22.9	44.4	67.3
	K_2CO_3	0.436	25.4	27.9	46.7	74.6
	$CaCl_2$	0.731	36.4	20.6	43.0	63.6
	NaCl	0.752	29.6	22.3	48.0	70.4
	$C_2H_3NaO_2$	0.819	32.6	23.0	44.4	67.4
	KCl[c]	0.809	33.8	24.4	41.8	66.2
		Average	32.9	23.2	43.9	67.1
		Standard Deviation	3.1	2.1	2.1	3.1

[c] Whole nut = shell + kernel.
[b] Kernels were moldy.
[c] Husks and kernels were moldy.

the Modified Henderson equation, the Modified Chung-Pfost equation, the Modified Halsey equation, the Modified Oswin equation, and a temperature-modified GAB equation (herein referred to as the Modified GAB equation) introduced by Jayas and Mazza (1993). This was done for each of the five categories (i.e., husk, shell, kernel, cluster, and whole nut) using non-linear least squares regression programs written specifically for this project. Results of these analyses are tabulated in table 3 for regressions with EMC as the dependent variable and in table 4 for regressions with ERH as the dependent variable.

With one exception, the five sorption isotherm equations used in this study are those highlighted and used in ASABE Standard D245.6. The one deviation is that ASABE Standard D245.6 lists the well-known Guggenheim-Anderson-deBoer (GAB) equation (herein referred to as the GAB model) and not a temperature-dependent form of the equation, as used in this study. Although in 1996 it was recommended that a temperature-modified form of the GAB equation be included in ASABE Standard D245.6 (Sokhansanj and Yang, 1996), this has not yet been done.

With EMC as the dependent variable, the GAB model ap-

Table 3. Desorption isotherm equation parameter estimates obtained with equilibrium moisture content as the dependent variable.

Sorption Isotherm Equation[a]	Category	Equation Parameter Estimate			SD[b]	MRD[c] (%)
		A	*B*	*C*		
Modified Henderson	Cluster	2.456E-04	1.470	68.39	0.96	5.6
	Husk	3.135E-04	1.215	84.89	2.27	9.0
$EMC = [\ln(1 - ERH) / (-A \times [T + C])]^{-B}$	Kernel	6.166E-04	1.782	56.77	0.49	5.9
	Whole nut	1.863E-04	1.700	59.63	0.74	4.9
	Shell	1.299E-04	1.698	55.35	0.89	4.9
Modified Chung-Pfost	Cluster	168.9	0.1357	38.5	0.77	3.3
	Husk	148.0	0.0950	43.4	2.42	7.6
$EMC = -\ln[-\ln(ERH) \times (T + C) / A] / B$	Kernel	223.4	0.4005	34.0	0.42	4.9
	Whole nut	198.8	0.1742	35.6	0.50	2.8
	Shell	187.6	0.1352	33.2	0.56	2.6
Modified Halsey	Cluster	4.730	-0.0138	2.036	0.75	5.1
	Husk	4.064	-0.0108	1.674	1.21	5.5
$EMC = [-\exp(A + B \times T) / \ln(ERH)]^{1/C}$	Kernel	3.609	-0.0160	2.420	0.45	6.0
	Whole nut	5.418	-0.0163	2.395	1.06	7.0
	Shell	6.032	-0.0174	2.393	1.34	7.2
Modified Oswin	Cluster	12.28	-0.07706	2.462	0.71	4.6
	Husk	14.14	-0.08498	2.017	1.56	6.8
$EMC = (A + B \times T) \times [ERH / (1 - ERH)]^{1/C}$	Kernel	5.206	-0.03180	2.948	0.44	5.5
	Whole nut	11.25	-0.07101	2.889	0.88	5.9
	Shell	14.53	-0.09657	2.886	1.09	6.0
Modified GAB	Cluster	7.912	0.7922	91.71	0.75	4.0
	Husk	7.752	0.8972	88.73	1.84	8.9
$EMC = (A \times B \times C \times ERH) / (T \times D \times E)$	Kernel	3.553	0.7173	120.5	0.45	6.1
where $D = 1 - B \times ERH$	Whole nut	7.817	0.7164	109.0	0.50	3.2
$E = 1 - B \times ERH + B \times C \times ERH/T$	Shell	10.17	0.7130	102.9	0.62	3.2

[a] ERH = equilibrium relative humidity (decimal), EMC = equilibrium moisture content (percent dry basis), and T = temperature (°C).
[b] SD = standard deviation = SQRT[residual sum of squares / (N – df)], where N =18, and df = degrees of freedom = 3.
[c] MRD (%) = mean relative deviation = $(100/N) \times \Sigma|(1 - \text{measured value} / \text{predicted value})|$.

Table 4. Desorption isotherm equation parameter estimates obtained with equilibrium relative humidity as the dependent variable.

Sorption Isotherm Equation[a]	Category	Equation Parameter Estimate			SD[b]	MRD[c] (%)
		A	*B*	*C*		
Modified Henderson	Cluster	2.871E-04	1.476	55.19	0.036	5.1
	Husk	2.301E-04	1.394	65.22	0.045	6.5
$ERH = 1 - \exp[-A \times (T + C) \times EMC^B]$	Kernel	7.584E-04	1.755	46.93	0.047	6.2
	Whole nut	2.966E-04	1.565	51.33	0.033	4.6
	Shell	2.163E-04	1.552	48.87	0.033	4.6
Modified Chung-Pfost	Cluster	183.2	0.1476	34.8	0.020	2.5
	Husk	190.6	0.1178	38.0	0.034	3.8
$ERH = \exp[-\exp(-B \times EMC) \times A / (T + C)]$	Kernel	247.1	0.4316	31.5	0.040	5.0
	Whole nut	191.0	0.1729	34.5	0.017	2.2
	Shell	180.8	0.1319	33.7	0.016	2.0
Modified Halsey	Cluster	4.263	-0.01970	1.817	0.032	4.0
	Husk	4.480	-0.01813	1.763	0.034	4.7
$ERH = \exp[-\exp(A + B \times T) / EMC^C]$	Kernel	3.316	-0.02081	2.203	0.041	5.4
	Whole nut	4.211	-0.01997	1.894	0.032	4.2
	Shell	4.599	-0.02035	1.860	0.035	4.9
Modified Oswin	Cluster	12.72	-0.1064	2.317	0.032	4.4
	Husk	15.41	-0.1219	2.175	0.038	5.6
$ERH = 1 / \{[(A + B \times T) / EMC]^C + 1\}$	Kernel	5.306	-0.0404	2.769	0.042	5.1
	Whole nut	11.25	-0.09336	2.447	0.031	4.0
	Shell	14.50	-0.1244	2.419	0.031	4.5
Modified GAB	Cluster	8.365	0.7838	74.53	0.034	4.6
	Husk	9.903	0.8182	70.77	0.434	5.6
$ERH = (2 + D - C/T - E^{0.5}) / [2 \times B \times (1 - C/T)]$	Kernel	3.601	0.7459	78.96	0.059	8.5
where $D = A \times C / (EMC \times T)$	Whole nut	7.493	0.7570	84.99	0.031	4.4
$E = 4 \times D - 2 \times D \times C/T + D^2 + (C/T)^2$	Shell	9.607	0.7465	97.49	0.025	3.5

[a] ERH = equilibrium relative humidity (decimal), EMC = equilibrium moisture content (percent dry basis), and *T* = temperature (°C).
[b] SD = standard deviation = SQRT[residual sum of squares / (*N* – df)], where *N* =18, and df = degrees of freedom = 3.
[c] MRD (%) = mean relative deviation = $(100/N) \times \Sigma|(1 - \text{measured value} / \text{predicted value})|$.

pears as:

$$EMC = (A \times B \times C \times ERH) / [(1 - B \times ERH)(1 - B \times ERH + B \times C \times ERH)] \quad (7)$$

where *A*, *B*, and *C* are parameters with physical meaning: *A* is the monolayer moisture content, *B* a parameter that takes into account the difference in chemical potential between the multilayers of adsorbed water and bulk water in the product, and *C* is an energetic constant also called the Guggenheim constant (Delgado and Sun, 2002). Note that equation 7 is obtained by simply striking the temperature (*T*) variable from the Modified GAB equation in tables 3 and 4. In short, the Modified GAB equation used in this study is the GAB model with parameter *C* replaced with the quantity *C*/*T*.

Equation 7 is commonly fit to individual sorption isotherm data, thereby producing different parameter estimates for each temperature value. When attempts were made to fit equation 7 to each of the 15 data sets (3 temperatures × 5 categories) in table 1, parameter *C* failed to converge (and instead approached infinity) during more than half of the regression analyses. Problems with parameter *C* approaching infinity were also reported by Chen and Jayas (1998) and are an indication of over-parameterization or of an inadequate or insufficient data set (Draper and Smith, 1981). In this study, the lack of convergence of parameter *C* was largely attributed to there being (1) only six points in each data set, and (2) insufficient differences or range in the ERH values of the six points. During individual regressions, essentially no change was observed in the residual sum of squares as estimates of parameter *C* diverged from a value of less than 100 to infinity, leading to the conclusion that parameter *C* should simply be taken as infinity, thereby reducing equation 7 to:

$$EMC = A / (1 - B \times ERH) \quad (8)$$

Chen and Jayas (1998) refer to equation 8 as the reduced form of the GAB model. Unlike the GAB model and Modified GAB, the reduced GAB does not force the isotherm through the origin.

The standard deviations and mean relative deviations from tables 3 and 4, along with residual plots and mean absolute residual values, were used to evaluate goodness-of-fit. Also investigated were differences between isotherms established using parameters obtained from the regression with EMC as the dependent variable and isotherms established using parameters obtained from the regression with ERH as the dependent variable. Ideally, these two sets of isotherms should plot right on top of each other because, in the end, they represent fits of the same equation to the same data set. However, because of the nonlinear nature of the isotherm equations with respect to all three variables, it is not uncommon for a slight change in a single parameter to increase the residual sum of squares with respect to one variable while simultaneously decreasing the residual sum of squares with respect to another variable. For lack of a better term, "isotherm mapping" or simply "mapping" herein refers to the similarity of isotherms generated using the same isotherm equation but with different sets of parameters. In this case, the two sets of parameters are those from regressions with EMC as the dependent variable and those from regressions with ERH as the dependent variable. Excellent mapping implies that the two sets of isotherms plot right on top of each other.

Table 5 contains the mean absolute residual values for EMC. A measureable difference in the mean absolute residual values for predictions obtained using the same equation with two different sets of parameters is an indication of poor isotherm mapping. For example, based on a survey of the values

Table 5. Mean absolute EMC residuals.

Sorption Isotherm Equation	Dependent Variable During Nonlinear Least Squares Regression[a]	Mean Absolute Residual for Predictions of EMC[b] (percent dry basis)				
		Husk	Kernel	Shell	Cluster	Whole Nut
Modified Henderson	ERH	1.68	0.36	0.85	0.78	0.66
	EMC	1.67	0.33	0.66	0.72	0.54
Modified Chung-Pfost	ERH	1.42	0.32	0.41	0.53	0.36
	EMC	1.55	0.29	0.41	0.51	0.35
Modified Halsey	ERH	1.26	0.40	1.42	1.07	1.00
	EMC	0.91	0.34	0.98	0.57	0.78
Modified Oswin	ERH	1.43	0.36	1.11	0.88	0.77
	EMC	1.18	0.31	0.80	0.52	0.64
Modified GAB	ERH	1.39	0.46	0.55	0.65	0.52
	EMC	1.43	0.33	0.45	0.53	0.38

[a] EMC = table 3 equation parameters used to predict EMC values; ERH = table 4 equation parameters used to predict EMC values.

[b] Mean absolute residual = $(1/N) \times \Sigma|$(measured value – predicted value)$|$.

in the cluster category of table 5, one may predict poor isotherm mapping for the Modified Halsey equation because of the significant difference in mean absolute residual values of 1.07% and 0.57%. When actually plotted, as shown in figure 4, this was indeed the case.

By every statistical measure, the Modified Chung-Pfost equation was found to provide the best overall fit to the experimental data for shells, kernels, clusters, and whole nuts. In the husk category, selection of the best-fitting model depends on the dependent variable used in the regression analyses. With EMC as the dependent variable, the Modified Halsey equation would be selected as the best fit for the husk category (table 3). With ERH as the dependent variable, the Modified Chung-Pfost equation would be selected as the best-fitting model (table 4). Figure 5 contains isotherm plots for the husk category generated using the Modified Chung-Pfost and Modified Halsey equations. Both plots show relatively poor isotherm mapping. This is a direct result of data points with low ERH values controlling the parameter estimates with ERH as the dependent variable, and data points with high ERH values controlling equation fits with EMC as the dependent variable. Not explored in this study was the impact on isotherm mapping of expressing EMC on a wet basis instead of a dry basis. As explained by Flood and White (1984), this switch is a non-linear transformation of the data that decreases the weight given to larger moisture content values and thus, in some cases, can reduce the standard error associated with equations that show greater lack of fit at higher EMC values. EMC was expressed on a dry basis in this study because all sorption isotherm model parameters given in ASABE Standard D254.6 were determined with EMC expressed on a dry basis.

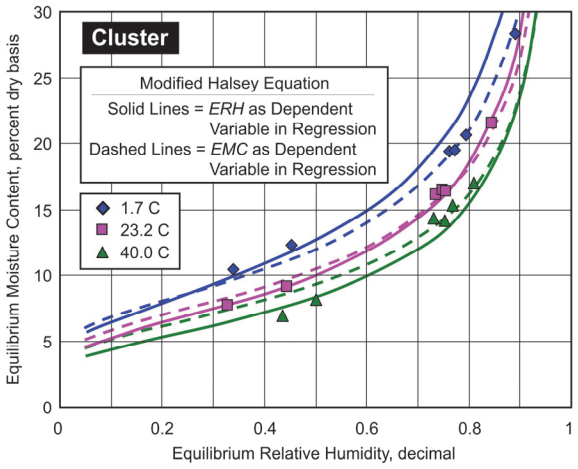

Figure 4. Differences in desorption isotherms for clusters resulting from a change in the dependent variable used in nonlinear least squares regressions involving the Modified Halsey equation.

The relative ability of the Modified Chung-Pfost equation to predict desorption behavior of hazelnut shells, kernels, clusters, and whole nuts is illustrated in figure 6. Note that the Modified Chung-Pfost equation in each plot is written with EMC as the dependent variable but contains parameters obtained from regressions in which ERH was the dependent variable. This selection (of parameter estimates from regressions with ERH as the dependent variable) was made because the mean relative deviations listed in table 4 were lower than those in table 3 for EMC as the dependent variable.

The Chung-Pfost equation plots for the husk category in figure 5 and for the cluster and whole nut categories in figure 6 show a poor fit to the data for ERH values above 0.80. For these higher ERH values, the Modified GAB equation showed an excellent fit in all five categories, as shown in figure 7, and is recommended for use in predicting EMC for ERH values above 0.70.

Moisture Content Differences

The data in table 1 and the isotherm plots show that husks, shells, and kernels have significantly different moisture desorption characteristics. This is fundamentally important to understand when specifying moisture contents for various operations. Under equilibrium conditions, the average kernel moisture content was found to be only 37% of that for shells, with a coefficient of variation (COV) of 9%. The EMC values for husks were on average 14% greater than those for shells, with a COV of 11%.

For safe storage of whole nuts, the moisture content of the kernels is of primary concern, but the moisture content of whole nuts is commonly measured for storage purposes. Conversely, when cracking nuts, the moisture content of the shell controls the cracking characteristics, but again, the moisture content of the whole nut is checked prior to cracking. For the 18 temperature and ERH combinations used in this study, the shell EMC averaged 128% (COV = 2.3%) of the whole nut EMC, whereas the kernel EMC averaged 47% (COV = 7.7%) of the whole nut EMC.

As previously explained, the lower EMC values for kernels can be attributed to their oil content, which Xu et al.

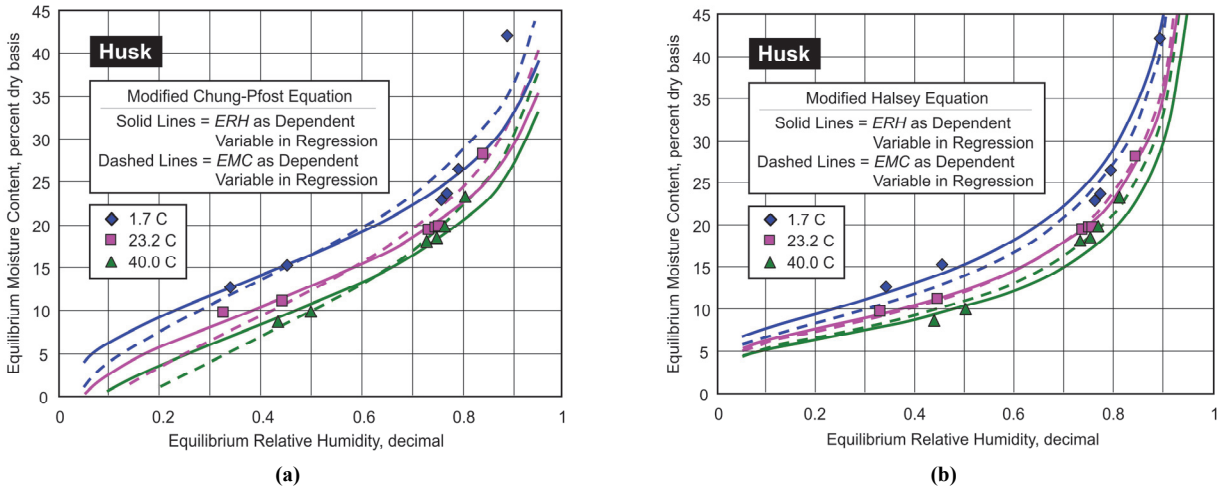

Figure 5. Differences in desorption isotherms for clusters resulting from a change in the dependent variable used in nonlinear least squares regressions involving the (a) Chung-Pfost equation and (b) Modified Halsey equation.

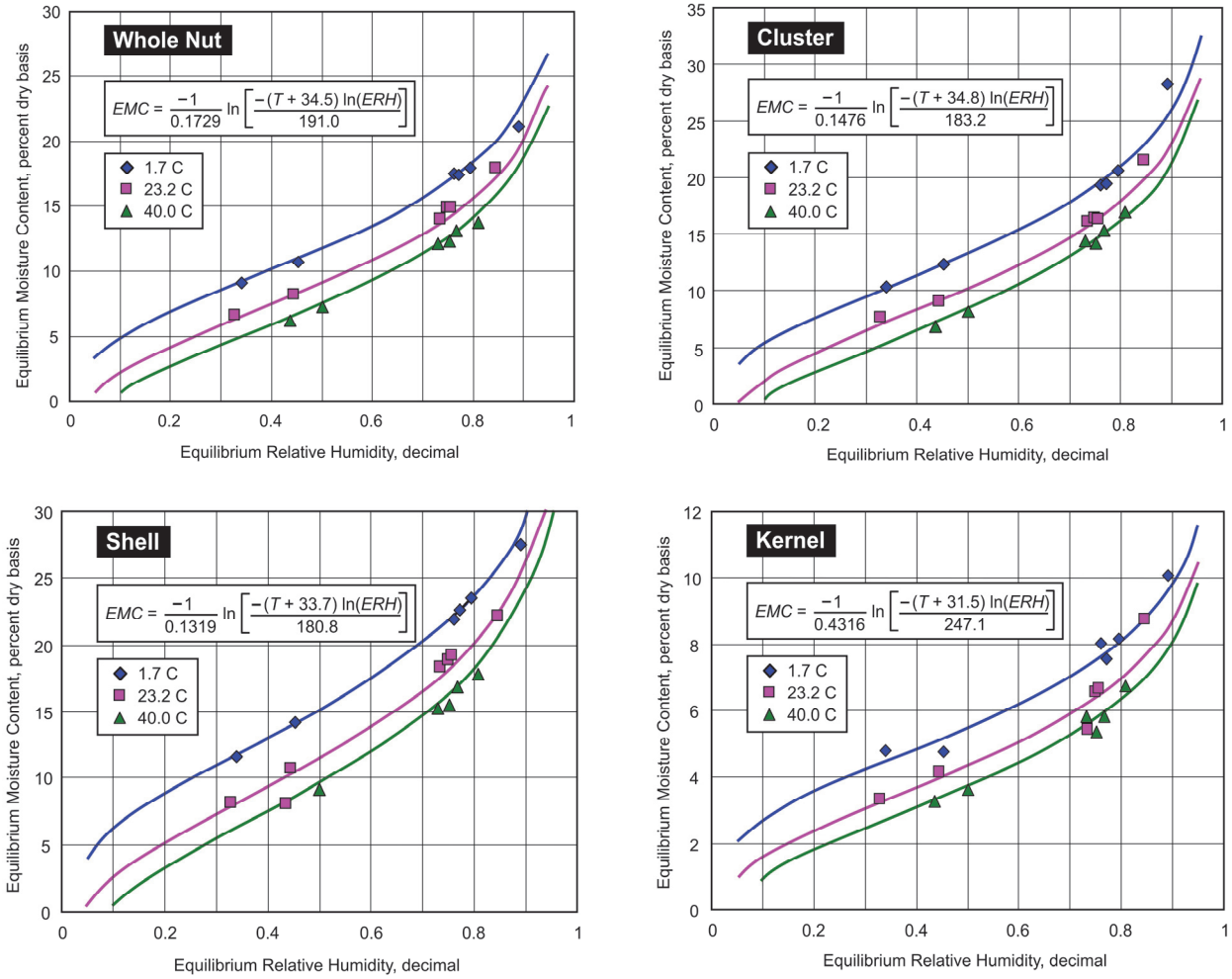

Figure 6. Experimental data and desorption isotherms at 1.7°C, 23.3°C, and 40.0°C obtained with the Modified Chung-Pfost equation using parameters from regression analyses with ERH as the dependent variable.

(2007) found to range from 51.4% to 75.1% of dry kernel weight for hybrid hazelnut genotypes with parentage similar to those investigated in this study. Xu et al. (2007) also reported that hazelnuts had an oil yield potential of 1000 kg ha^{-1} (about twice the oil yield potential of soybeans), that hazelnut oils are unique in that only two fatty acids (oleic and linoleic) account for 90% of their fatty acid content, and that the high level of oleic and linoleic acids, combined with a low linolenic acid content, improve the thermo-oxidative stability of hazelnut oils, while low levels of saturated (palmitic and

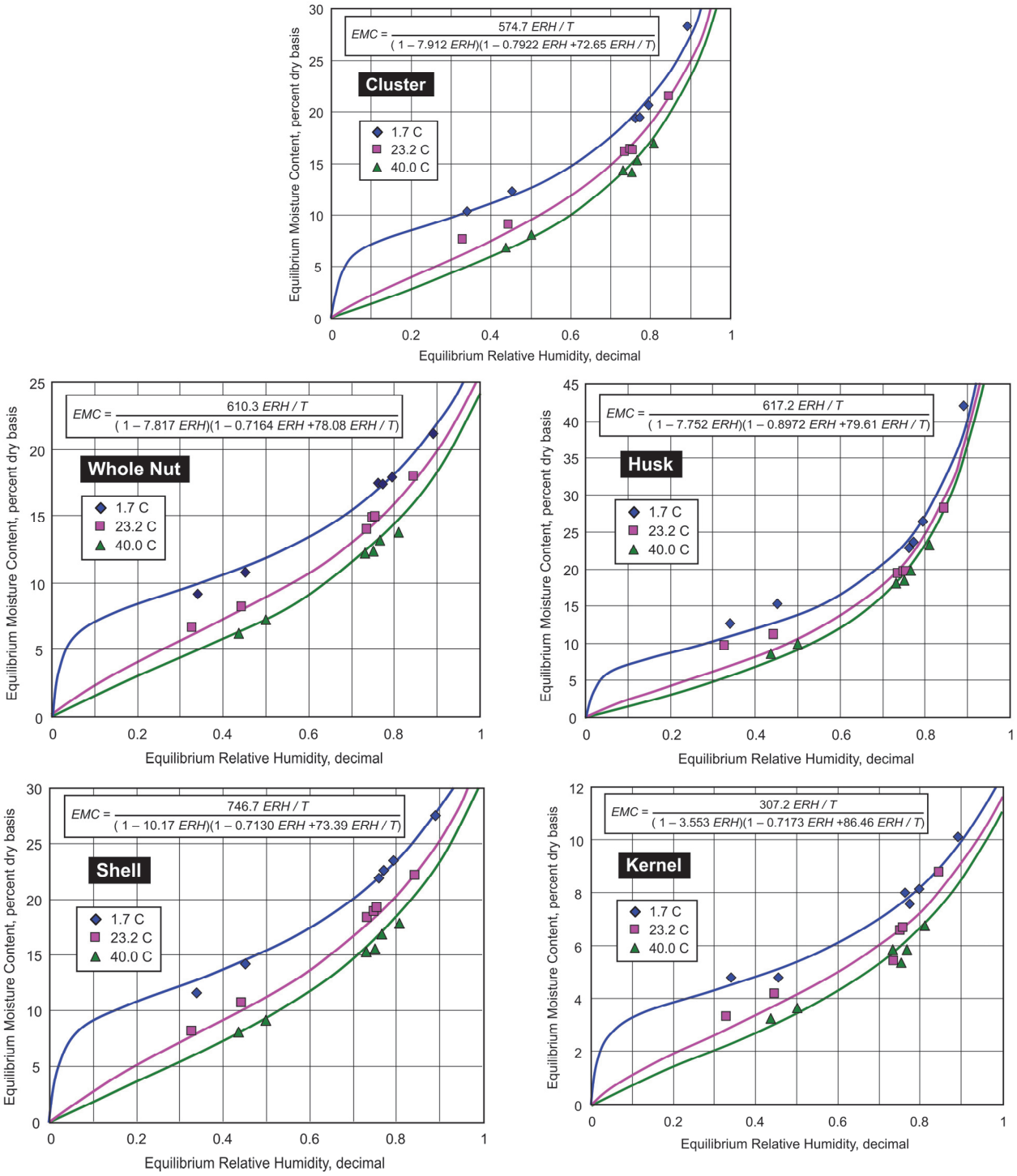

Figure 7. Experimental data and desorption isotherms at 1.7°C, 23.3°C, and 40.0°C obtained with the Modified GAB equation.

stearic) acids enhance the properties of the oils in cold environments. Because of these and other unique characteristics, kernel oil content will be a major consideration when making hybrid hazelnut germplasm selections for wide-scale cultivation of hazelnuts in the Upper Midwest. Although kernel oil content was not measured as part of this study, it should be part of future investigations of kernel EMC, especially after hybrid hazelnut varieties become established and the food safety during storage of these varieties becomes more critical. Also of interest may be the extent to which the oil content varies from kernel to kernel within a particular plant and from plant to plant within a particular variety, as well as the extent to which the oil content and composition are affected by harvesting clusters in a green state instead of allowing nuts to fully ripen on the plant.

Mass Ratios

Mass ratios are essential for two reasons: (1) they are needed to predict kernel yield from whole nut yields or known cluster yields, and (2) they are required when calculating clus-

ter and whole nut EMC values from component EMC values, as was done in this study. Figure 8 shows the mass ratios for cluster fractions and whole nut fractions. On a dry matter basis, the average cluster mass was 32.9% husk, 43.9% shell, and 23.2% kernel (table 2). Likewise, on a dry matter basis, the average whole nut mass was 65.5% shell and 34.5% kernel. As the moisture content of clusters and whole nuts increases, the percentage of their total mass that is kernels will decrease due to the relatively greater amount of moisture gained by the husks and shells. For whole nuts, the decrease in percent kernel mass averaged 1.6% for every 10% increase in whole nut moisture content.

The values in table 2 are the average mass ratios for each of the 18 aggregate samples conditioned in this study. The variability in the kernel content of individual whole nuts is show in figure 9. This distribution was obtained from measurements made on 350 whole nuts randomly selected from the six biomaterial pails maintained at 23.3°C. On average, 32.7% of the mass of an undried whole nut was kernel, with a standard deviation of 5.4%. The 32.7% average is lower than the 35% mean reported by Braun et al. (2019) for the 75 specially selected Upper Midwest grown hybrid hazel genotypes that they studied. Braun et al. (2019) also reported an average whole nut kernel composition of 38% for their top eight genotypes, with the highest at 42%. This value is still significantly lower than that for most *C. avellana* varieties, which can approach 50%. The same research also reported that, within a specific genotype, the standard deviation of whole nut kernel composition was 1% to 2%.

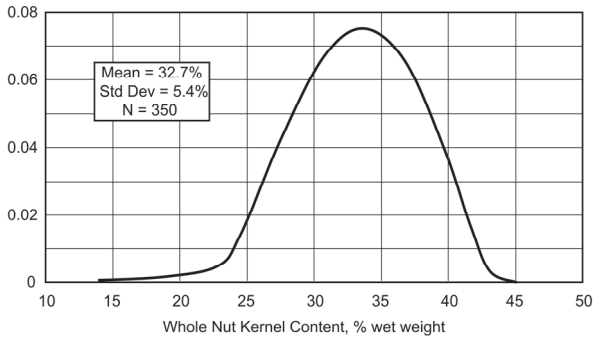

Figure 9. Distribution of individual whole nut kernel content.

Summary and Conclusions

As part of an effort to develop a thriving hazelnut industry in the Upper Midwest, researchers at the University of Wisconsin-Madison have been developing specialized hazelnut harvesting and processing techniques and equipment. This study was undertaken because proper assessment and optimization of this equipment requires knowledge of the EMC properties of the hybrid hazelnuts (*C. americana* × *C. avellana*) currently being cultivated in the Upper Midwest.

For this study, hazelnut clusters were hand-picked from a research plot in Stoughton, Wisconsin, and dried in 18 different controlled environments (six different RH levels at three different temperatures). Actual moisture conditioning took place over saturated salt solutions in specially fabricated biomaterial moisture conditioning units (Bohnhoff, 2017).

After a six-week period during which the clusters reached equilibrium with their environment, the clusters were separated into husk, shell, and kernel fractions and returned to their respective conditioning units. After another six weeks in the conditioning units, the moisture content of each fraction was determined by oven-drying at 103°C for 48 h.

Significant differences were found between the EMC values of husks, shells, and kernels. Under equilibrium conditions, the average kernel moisture content was found to be only 37% of that for shells, whereas the EMC values for husks were on average 14% greater than those for shells.

Experimental EMC, ERH, and temperature data were fit to the Modified Henderson equation, Modified Chung-Pfost equation, Modified Halsey equation, Modified Oswin equation, and a temperature-modified GAB equation (herein referred to as the Modified GAB equation). Although the Modified Chung-Pfost equation was found to provide the best overall fit to the experimental data, the Modified GAB equation is recommended for predicting desorption behavior for ERH values above 0.70.

Mass ratios were also calculated. On a dry basis, the average cluster mass was 32.9% husk, 43.9% shell, and 23.2% kernel. Likewise, on a dry basis, the average whole nut mass was 65.5% shell and 34.5% kernel. As the moisture content of clusters and whole nuts increases, the percentage of their total

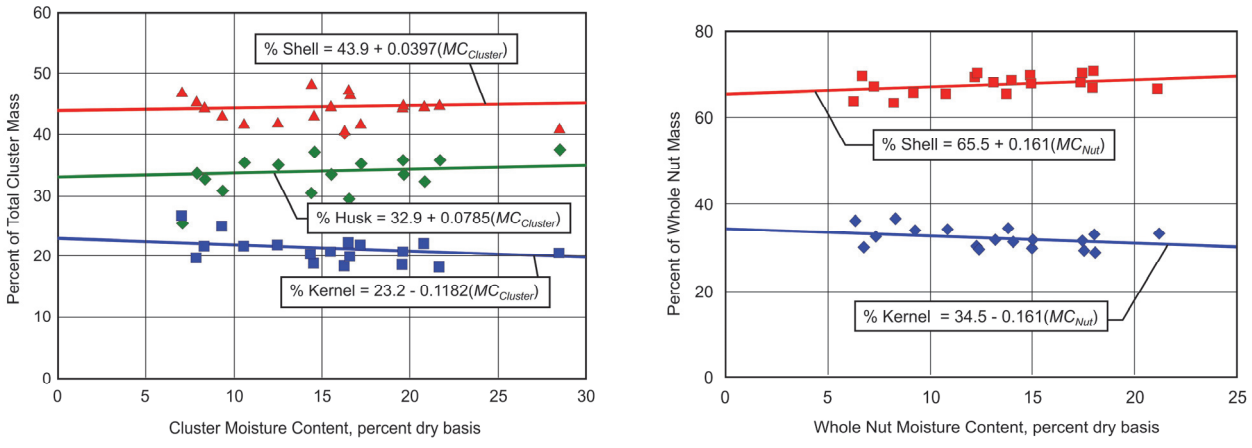

Figure 8. Mass ratios for (a) cluster fractions and (b) whole nut fractions.

mass that is kernels will decrease due to the relatively greater amount of moisture gained by the husks and shells.

Overall, the biomaterial moisture conditioning system used in this study worked very well, and thus it is recommended for use by others.

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