

1 Introduction: Historical Highlights of Water Activity Research

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The concept of water activity (a_w) is more than 50 years old. William James Scott showed in 1953 that microorganisms have a limiting a_w level for growth. It is now generally accepted that a_w is more closely related to the microbial, chemical, and physical properties of foods and other natural products than is total moisture content. Specific changes in color, aroma, flavor, texture, stability, and acceptability of raw and processed food products have been associated with relatively narrow a_w ranges (Rockland and Nishi 1980). Next to temperature, a_w is considered one of the most important parameters in food preservation and processing (van den Berg 1986). This chapter is not a review of the literature on a_w but rather a highlight of some early key a_w research as it relates to microbial growth, moisture sorption isotherms, prediction and measurement of a_w in foods, and, to a lesser extent, the influence of a_w on the physical and chemical stability of foods.

Australian-born microbiologist Scott (1912–1993) received his bachelor's degree from the University of Melbourne (1933) and a doctorate of science degree from the Council for Scientific and Industrial Research (CSIR) Meat Research Laboratory (1933). He then took a position as senior bacteriologist at the CSIR Division of Food Preservation and Transport from 1940 to 1960. In 1960, he moved to the Meat Research Laboratory, where he served as assistant chief of division until 1964 and officer-in-charge until 1972. In 1979, he became a fellow of the Australian Academy of Technological Sciences and Engineering.

Scott's early work was concerned with handling, cooling, and transport conditions that would enable chilled beef to be successfully exported to Britain. During World War II, he was concerned with the microbiology of foods supplied by Australia to the Allied Forces. After the war, he pioneered studies on the water relations of microorganisms. In 1953, Scott related the relative vapor pressure of food to the thermodynamic activity of water, using the definition $a_w = p/p_o$, where a_w is the water activity derived from the laws of equilibrium thermodynamics, p is the vapor pressure of the sample, and p_o is the vapor pressure of pure water at the same temperature and external

Table 1.1 Papers by Scott and Christian.

Author	Year	Title of Paper
Scott, W.J.	1953	Water relations of <i>Staphylococcus aureus</i> at 30°C
Christian, J.H.B. and Scott, W.J.	1953	Water relations of <i>Salmonella</i> at 30°C
Christian, J.H.B.	1955a	The influence of nutrition on the water relations of <i>Salmonella oranienburg</i>
Christian, J.H.B.	1955b	The water relations of growth and respiration of <i>Salmonella oranienburg</i> at 30°C
Scott, W.J.	1957	Water relations of food spoilage microorganisms

pressure. He showed a clear correlation between the a_w of the growth medium and the rate of *Staphylococcus aureus* growth. The summary of his paper stated:

Fourteen food-poisoning strains of *Staphylococcus aureus* have been grown in various media of known a_w at 30°C. Aerobic growth was observed at water activities between 0.999 and 0.86. The rate of growth and the yield of cells were both reduced substantially when the a_w was less than 0.94. The lower limits for growth in dried meat, dried milk, and dried soup were similar to those in liquid media. Aerobic growth proceeded at slightly lower water activities than anaerobic growth. All cells were capable of forming colonies on agar media with water activities as low as 0.92. The 14 strains proved to be homogeneous with similar water requirements.

Scott's classic demonstration that it is not the water content but the a_w of a food system that governs microbial growth and toxin production was a major contribution to food microbiology. Many scientists, most notably his Australian colleague, J.H.B. Christian, expanded Scott's work. Key papers published in the 1950s by both Scott and Christian are listed in Table 1.1. These papers laid the foundation for future research into the survival and growth of microorganisms in foods at low a_w .

In the field of food science, the general acceptance and application of the concept of a minimum a_w for microbial growth began with the review by Scott published in 1957, *Water Relations of Food Spoilage Microorganisms*. Taken from the table of contents in Scott's classic review, the following are some of the aspects discussed:

- III. Methods for controlling a_w :
 - Equilibration with controlling solutes
 - Determination of the water sorption isotherms
 - Addition of solutes
- IV. Water requirements for growth
 - Molds
 - Yeasts
 - Bacteria
 - General relationships
- V. Factors affecting water requirements
 - Nutrition, temperature, oxygen, inhibitors, adaptation
- VI. Special groups
 - Halophilic bacteria
 - Osmophilic yeasts
 - Xerophilic molds
- VII. Some applications in food preservation
 - Fresh foods, dried foods, concentrated foods, frozen foods, canned foods

Table 1.2 Selected early work on the minimal water activity for growth of pathogenic and spoilage microorganisms.

Author	Year	Title of Paper
Baird-Parker, A.C. and Freame, B.	1967	Combined effect of water activity, pH, and temperature on the growth of <i>Clostridium botulinum</i> from spore and vegetative cell inocula
Ohye, D.F. and Christian, J.H.B.	1967	Combined effects of temperature, pH, and water activity on growth and toxin production by <i>Clostridium botulinum</i> types A, B, and E
Pitt, J.I. and Christian, J.H.B.	1968	Water relations of xerophilic fungi isolated from prunes
Anand, J.C. and Brown, A.D.	1968	Growth rate patterns of the so-called osmophilic and non-osmophilic yeasts in solutions of polyethylene glycol
Ayerst, G.	1969	The effects of moisture and temperature on growth and spore germination in some fungi
Emodi, A.S. and Lechowich, R.V.	1969	Low temperature growth of type E <i>Clostridium botulinum</i> spores. II. Effects of solutes and incubation temperature
Kang, C.K., Woodburn, M., Pagenkopf, A., and Cheney, R.	1969	Growth, sporulation, and germination of <i>Clostridium perfringens</i> in media of controlled water activity
Horner, K.J. and Anagnostopoulos, G.D.	1973	Combined effects of water activity, pH, and temperature on the growth and spoilage potential of fungi
Troller, J.A.	1972	Effect of water activity on enterotoxin A production and growth of <i>Staphylococcus aureus</i>
Beuchat, L.R.	1974	Combined effects of water activity, solute, and temperature on the growth of <i>Vibrio parahaemolyticus</i>
Northolt, M.D., van Egmond, H.P., and Paulsch, W.W.	1977	Effect of water activity and temperature on aflatoxin production by <i>Aspergillus parasiticus</i>
Pitt, J.I. and Hocking, A.D.	1977	Influence of solute and hydrogen ion concentration on the water relations of some xerophilic fungi
Lotter, L.P. and Leistner, L.	1978	Minimal water activity for enterotoxin A production and growth of <i>Staphylococcus aureus</i>
Hocking, A.D. and Pitt, J.I.	1979	Water relations of some <i>Penicillium</i> species at 25°C
Briozzo, J., de Lagarde, E.A., Chirife, J., and Parada, J.L.	1986	Effect of water activity and pH on growth and toxin production by <i>Clostridium botulinum</i> type G
Tapia de Daza, M.S., Villegas, Y., and Martinez, A.	1991	Minimal water activity for growth of <i>Listeria monocytogenes</i> as affected by solute and temperature

Since the work of Scott, a_w has become one of the most important intrinsic properties used for predicting the survival and growth of microorganisms in food, due to its direct influence on product stability and quality. Thus, the minimal a_w level for growth emerged as one of the most investigated parameters for determining the water relations of microorganisms in foods. This limiting value defines, in theory, the level below which a microorganism or group of microorganisms can no longer reproduce. The limiting value will not be the same for all microorganisms, and some may be able to tolerate low a_w , and still compromise product safety. The understanding and control of a_w contributes to safer food storage conditions in general and forms the basis of much modern food formulation, especially for intermediate-moisture foods.

Several workers developed studies to determine the minimal a_w level for growth of bacterial pathogens, yeasts, and molds and the production of microbial toxins. Table 1.2 displays some selected papers by authors following the work of Scott and Christian. It is worth mentioning that in 1978, Troller and Christian published a book on a_w entitled *Water Activity and Food*.

In addition to the experimental determination of minimal a_w for microbial growth, researchers also were concerned with the mechanism of cell adaptation to low a_w ,

Table 1.3 Low water activity adaptation.

Author	Year	Title of Paper
Christian, J.H.B. and Waltho, J.A.	1961	The sodium and potassium content of nonhalophilic bacteria in relation to salt tolerance
Christian, J.H.B. and Waltho, J.A.	1962	The water relations of staphylococci and micrococci
Christian, J.H.B. and Waltho, J.A.	1964	The composition of <i>Staphylococcus aureus</i> in relation to the water activity of the growth medium
Brown, A.D., and Simpson, JR.	1972	Water relations of sugar tolerant yeasts: the role of intracellular polyols
Gould, G.W and Measures, J.C.	1977	Water relations in single cells
Brown, A.D.	1975	Microbial water relations. Effect of solute concentration on the respiratory activity of sugar tolerant and non-tolerant yeasts
Measures, J.C.	1975	Role of amino acids in osmoregulation of nonhalophilic bacteria
Chirife, J., Ferro Fontán, C., and Scorza, O.C.	1981	The intracellular water activity of bacteria in relation to the water activity of the growth medium
Anderson, C.B. and Witter, L.D.	1982	Glutamine and proline accumulation by <i>Staphylococcus aureus</i> with reduction in water activity

specifically, the intracellular composition of cells grown at reduced a_w . Some key papers discussing low a_w adaptation are listed in Table 1.3.

Two important aspects related to microbial water relations – the solute effects and the influence of a_w on the thermal resistance of microorganisms, specifically – were the subject of early studies by various researchers. Table 1.4 shows a compilation of some classic papers on a_w and microbial water relations.

In addition to the research on a_w and microbial control, during the 1960s and 1970s, information about a_w and its influence on the chemical, enzymatic, and physical stability of foods began to appear rapidly in the literature. Research was conducted on the influence of a_w to (i) control undesirable chemical reactions, (ii) prolong the activity of enzymes, (iii) understand the caking and clumping of powders, and (iv) optimize the physical properties of foods such as texture and moisture migration.

Labuza (1970) presented a comprehensive review on the influence of a_w on chemical reactions in foods. Since then, extensive studies have been conducted in this area and are reviewed in Duckworth (1975), Labuza (1980), Rockland and Nishi (1980), Rockland and Stewart (1981), and Leung (1987). The a_w of a food describes the energy status of the water in that food and, hence, its availability to act as a solvent and participate in chemical or biochemical reactions (Labuza 1977). The ability of water to act as a solvent, medium, and reactant increases as a_w rises (Labuza 1975). Water activity influences nonenzymatic browning, lipid oxidation, degradation of vitamins, and other degradative reactions. The influence of a_w on the rate of nonenzymatic browning reactions, also called Maillard reactions, is described by Troller and Christian (1978b), Labuza and Saltmarch (1981), Nursten (1986), and Bell (1995). The influence of a_w on lipid oxidation has been studied extensively and reviewed by Labuza (1975), Troller and Christian (1978a), and (Karel and Yong 1981; Karel 1986).

Enzyme activity and stability are influenced significantly by a_w due to their relatively fragile nature (Blain 1962; Acker 1969; Potthast et al. 1975; Potthast 1978; Schwimmer 1980; Drapron 1985). Most enzymes and proteins must maintain conformation to remain active. Maintaining critical a_w levels to prevent or entice conformational changes in enzymes is important to food quality. Most enzymatic reactions are slowed down at water activities below 0.80, but some reactions occur even at very low a_w values.

Table 1.4 Microbial water relations.

Author	Year	Title of Paper
Marshall, B.J., Ohye, D.F., and Christian, J.H.B.	1971	Tolerance of bacteria to high concentrations of NaCl and glycerol in the growth medium
Baird-Parker, A.C., Boothroyd, M., and Jones, E.	1970	The effect of water activity on the heat resistance of heat-sensitive and heat-resistant strains of <i>Salmonellae</i>
Horner, K.J. and Anagnostopoulos, G.D.	1975	Effect of water activity on heat survival of <i>Staphylococcus aureus</i> , <i>Salmonella typhimurium</i> , and <i>Salmonella steffenberg</i>
Corry, J.E.L.	1976	The effect of sugars and polyols on the heat resistance and morphology of osmophilic yeasts
Goepfert, J.M., Iskander, I.K., and Amundson, C.H.	1970	Relation of the heat resistance of <i>Salmonellae</i> to the water activity of the environment
Jakobsen, M. and Murrel, W.G.	1977	The effect of water activity and a_w -controlling solute on sporulation of <i>Bacillus cereus</i> T
Christian, J.H.B.	1981	Specific solute effects on microbial water relations

Water activity affects the stability, flow, and caking and clumping of powders during storage (Peleg and Mannheim 1977; Saltmarch and Labuza 1980; Chuy and Labuza 1994; Aguilera and del Valle 1995). Controlling a_w in a powder product below critical levels maintains proper product structure, texture, flowability, density, and rehydration properties. Knowledge of the a_w of powders as a function of moisture content and temperature is essential during processing, handling, packaging, and storage to prevent the deleterious phenomenon of caking, clumping, collapse, and stickiness. Caking is dependent on a_w , time, and temperature and is related to the collapse phenomena of the powder under gravitational force (Chuy and Labuza 1994).

Water activity affects the textural properties of foods (Troller and Christian 1978a; Bourne 1987, 1992). Foods with high a_w have a texture that is described as moist, juicy, tender, and chewy. When the a_w of these products is lowered, undesirable textural attributes such as hardness, dryness, staleness, and toughness are observed. Food low in a_w normally have texture attributes described as crisp and crunchy, while at higher a_w , the texture becomes soggy. The crispness intensity and overall hedonic texture of dry snack food products are a function of a_w (Katz and Labuza 1981; Hough et al. 2001). Critical water activities are found where the product becomes unacceptable from a sensory standpoint. Glass transition theory from the study of polymer science aids in understanding textural properties and explains the changes that occur during processing and storage (Sperling 1986; Roos and Karel 1991; Roos 1993; Slade and Levine 1995). Physical structure is often altered by changes in a_w due to moisture gain, resulting in a transition from the glassy to the rubber state.

With the introduction of the concept of a_w , it is possible to describe the relationship between a_w and food moisture content, i.e. the moisture sorption isotherm. Notable among the first published papers on water sorption isotherms of foods are the works of Makower and Dehority (1943) on dehydrated vegetables, Makower (1945) on dehydrated eggs, and Gane (1950) on fruits and vegetables. It is interesting to note that these authors made references to “equilibrium relative humidity” or “relative vapor pressure” instead of “water activity.”

A variety of mathematical models have been developed to describe the typical sigmoidal moisture sorption isotherm of foods. However, before the advent of computers

and the availability of nonlinear regression software, it was necessary to use two-parameter models that could be transformed into a linear equation, from which the fitting parameters could be determined. Notable among two-parameter models is the Brunauer–Emmett–Teller (BET) equation (see Appendix C). Pauling (1945) applied the BET equation to water sorption by proteins to correlate the “monolayer value” with the number of polar groups. Additionally, the empirical models developed by Oswin (1946) and Henderson (1952) were extensively used for food isotherm development.

Many multicomponent foods contain ingredients that have different water activities, and during storage, moisture will be exchanged until a final equilibrium a_w is reached. Salwin and Slawson (1959) developed a simple and useful equation to predict that equilibrium a_w . Later, Labuza (1968) published a classic paper entitled “Sorption Phenomena of Foods,” which reviewed the main concepts of water sorption phenomena in foods as well the most popular sorption models being used. In 1969, Rockland applied Henderson’s equation to sorption data in several foods and introduced the idea of “localized isotherms.” Localized isotherms divide the curve into three regions separated by intercepts that delineate major differences in the type and character of the water binding in the system. Iglesias et al. (1975), Iglesias and Chirife (1976) showed that a multilayer adsorption equation, originally developed by Halsey for physical adsorption on nonuniform surfaces, could be used (reasonably well) to describe the water sorption isotherms of a great variety of foods and food components. This equation became one of the most successful two-parameter models for describing the sorption behavior of foods.

Chirife and Iglesias (1978) published a review of literature on equations for fitting water sorption isotherms of foods and food products. At that time, they were able to compile 23 equations for correlating equilibrium moisture content in food systems. Compilations of moisture sorption isotherms for a large number of foods and food components were published by Wolf et al. (1973) and Iglesias and Chirife (1982).

At present, the most popular sorption isotherm model in the food area is the Guggenheim–Anderson–de Boer (GAB) equation (see Appendix C). It is an extension of the BET equation, but with an additional parameter. Van den Berg (1981) and Bizot (1983) were among the first to demonstrate that the sigmoid-shaped isotherms of food could be precisely fitted up to about 0.90 a_w using the GAB equation. Since then, several key papers have been published to further corroborate the goodness of fit of the GAB equation (Lomauro et al. 1985a, 1985b; Iglesias and Chirife 1995).

The first attempts to predict the a_w in food solutions were in confectionery products. Grover (1947) developed an empirical method to predict the a_w of confectionery solutions. He reported a relationship between the concentrations of solutions of different sugars having the same a_w . Money and Born (1951) also proposed an empirical equation for calculating a_w in solutions of sugars and sugar syrups. In contrast to Grover or Money and Born, Norrish (1966) did not propose an empirical relationship, but derived a model for predicting the a_w of nonelectrolyte solutions based on the laws of thermodynamics. This is probably the most commonly used model for predicting a_w in nonelectrolyte binary solutions, due to availability of the parameter (constant K) needed for predictions. In the 1970s and 1980s, interest in controlling a_w in intermediate-moisture foods stimulated research in the prediction of the a_w of single and mixed nonelectrolyte solutions (Bone 1973; Chuang and Toledo 1976). Kaplow (1970)

described the use of Raoult's law to calculate a_w in intermediate-moisture foods. Chirife et al. (1980) continued Norrish's work and reported values of the parameter K for a wide variety of nonelectrolytes (food solutes). Karel (1973) published an update on recent research and development in the field of low- and intermediate-moisture foods, including moisture sorption and a_w in foods.

Perhaps one of the most useful prediction equations in the food area is the Ross equation. Ross (1975) published his very simple equation for estimating the a_w of multicomponent solutions, even highly concentrated food solutes, which proved to be useful for most a_w predictions. In 1981, Ferro Fontán and Chirife developed a refinement of the Ross equation, which allowed a better estimate of a_w of multicomponent solutions.

As knowledge about the importance of a_w increased, food scientists needed a better measurement to quantify a_w . Many early methods for the measurement of a_w of foods were adaptations of atmospheric humidity measurement techniques. There are a number of excellent reviews on a_w measurement (Smith 1971; Labuza et al. 1976; Troller and Christian 1978c; Rizvi 1986; Wiederhold 1987; Fontana and Campbell 2004). Stoloff (1978) reported the results of a collaborative study on the calibration of a_w measuring instruments and devices. Today, there are several commercially available a_w meters that allow rapid, accurate, and reproducible measurement.

In conclusion, since Scott's early work, an enormous pool of basic information about a_w and its relation to the safety, quality, and stability of foods has been generated. This increased knowledge in the understanding of a_w has led to numerous intermediate-moisture food products being developed commercially. Since the early 1960s, a large number of patents describing practical applications of a_w in foods have been issued in the United States and internationally (Bone 1987). Water activity technology also aids in the development of nutritious shelf-stable food for the National Aeronautics and Space Administration (NASA) and the US military in the Meals Ready-to-Eat (MRE) program. Future research and application of a_w will aid food scientists in the development of new foods that are safe, shelf-stable, easy to prepare, and highly nutritious.

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